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**SORPTION AND DIFFUSION STUDIES
IN CEMENTITIOUS GROUTS
I: CURING SOLUTION - SCSS**

Report No. 91-164-K

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ABSTRACT

Laboratory experiments were conducted to evaluate the contaminant transport properties of cement-based sealing materials for use in vault sealing and fracture grouting within an underground repository for nuclear fuel waste. Diffusion coefficients were measured for tritiated water and ^{36}Cl under simulated saline groundwater conditions for six mixtures of Sulphate Resistant Portland Cement and Silica Fume at two water to cement ratios. Tortuosity factors were calculated from the results of the diffusion tests and retardation factors for ^{137}Cs and ^{89}Sr were estimated from measured distribution coefficient, bulk density and porosity values.

The results of the diffusion experiments show both tritiated water and ^{36}Cl to behave as reactive tracers in cement-based materials. The water to cement ratio was shown to have a significant effect on the retardation capabilities of the grout mixes with retardation factors increasing as the water to cement (w/c) ratio was reduced from 0.35 to 0.25. The results also showed the addition of silica fume to have impact on the potential migration rates of both ^{137}Cs and ^{89}Sr , however, under saline conditions, a similar reduction in the effective diffusion coefficients could be achieved through a decrease in the water to cement ratios.

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SORPTION AND DIFFUSION STUDIES IN CEMENTITIOUS GROUTS

1.0 INTRODUCTION

The Canadian concept for nuclear fuel waste disposal focuses on disposal in deep underground repositories constructed in a granitic pluton in the Canadian Shield. In addition to the inherent fracturing in these rocks, further fracturing may be induced during the excavation process. These fractures provide a preferential pathway for groundwater flow and, therefore, radionuclide migration. Currently both cementitious and clay-based materials are being considered for use in vault sealing and fracture grouting. It is therefore important to determine the contaminant transport properties of these candidate materials.

As part of the Technical Assistance Program to AECL (TAP), the contaminant transport properties and retardation capabilities of selected cement-based grout materials were evaluated. Specifically, the adsorption of strontium (Sr) and cesium (Cs) and the diffusion of tritiated water (^3H) and ^{36}Cl ions were studied in cement pastes comprised of Sulphate Resistant Portland cement (Canadian Type 50, ASTM Type V) blended with various amounts of silica fume (SF) and cured using solutions of varying ionic strengths. From the diffusion experiments the physical transport properties of the various cement grouts were evaluated by a comparison of the respective tortuosity factors. Cesium and strontium were chosen as an initial means of comparing the radionuclide retardation capabilities of the mixes due to their relatively simple solution chemistries. In addition to the grout samples, Cs and Sr distribution coefficients and non-reactive (^3H and ^{36}Cl) diffusion coefficients were measured for granite samples obtained from the Lac du Bonne batholith. In this phase of the investigation, the grout specimens were cured and all experiments conducted in a simulated saline groundwater solution (Standard Canadian

Shield Saline Solution). Additional experiments are being conducted using a simulated granitic groundwater (WN-1) and saturated limewater solutions. Diffusion experiments using the WN-1 solution are being conducted both at room temperature and 60 °C.

2.0 MATERIALS AND METHODS

2.1 Sample Description and Preparation

2.1.1 Grout Mixes

Six cement grout mixes were studied (Table 1). Type 50 Sulphate Resistant Portland cement was blended with silica fume (fine-grained amorphous silica) using Mighty 150, a water reducing superplasticizer (naphthalene sulphonate formaldehyde condensate). The proportion of water (water to solids ratio (w/c)) was also modified to analyze its effect on diffusion and adsorption.

The grout samples were mixed according to the proportions given in Table 1, cast in 5 cm (2") by 10 cm (4") brass cylinders and allowed to cure for 24 hours at 100% humidity prior to placement in the curing solution. The samples were cured in Standard Canadian Shield Saline solution (SCSS) for 56 days before testing. For each of the mix formulations, six samples were cast and cured. The chemical composition of the curing solution prior to and after contact with the cement grouts are shown in Table 2.

After the curing period, discs were cut from each of the mix formulations for use in the diffusion experiments. Before slicing, precipitated salts deposited on the surface of the grout castings were removed.

Prior to processing, the top 3 mm of each cylinder was discarded. In addition, a further 3 cm from grout samples 13960 and 13963 were removed due to its mottled, inhomogeneous appearance. Each grout cylinder was sliced using a diamond rock saw equipped with a 20 cm (8") diameter blade into approximately 8 mm slabs for use in the diffusion experiments. In order to attain a smooth surface and uniform thickness, each disc was ground to 5.9 mm using a Do All Model #VS612 surface grinder with a 180 grit diamond wheel. The discs were rinsed with deionized water and placed in glass beakers filled with SCSS solution and placed in a desiccator under vacuum to ensure that saturation was maintained. Before being used in the diffusion experiments the physical measurements of each disc were recorded (diameter, thickness, surface area and volume) and are presented in Appendix A.

Table 1: Physical Characteristics of Samples

Percent T50 Cement	Percent Silica Fume	Percent Superplasticizer	W/C	Sample No.	Bulk Density (g/cm ³)	Porosity*
100	---	1	0.35	13960	1.99	0.181
			0.25	13963	2.17	0.113
90	10	1	0.35**	13959	1.71	0.225
		3	0.25	13962	1.94	0.142
85	15	1	0.35	13961	1.68	0.198
		3	0.25	13964	1.88	0.130
Lac du Bonne Granite		--	--	--	2.60	0.004

* determined by mercury porosimetry
** AECL reference grout

Table 2: Standard Canadian Shield Saline Solution
Chemical Analyses of Curing Solutions

	Initial Solution Concentration (mg/L)	After Curing Period Concentration (mg/L)
Ca	15800	11000
Mg	220	0.43
Na	5200	5400
K	52	1100
Sr	35	40
SO ₄	600	<600
Cl	37000	30000
TIC	<0.5	3.5
pH	5.9	12.5

The remaining grout not used for diffusion testing was crushed for use in the equilibrium batch experiments. For the determination of distribution coefficients (K_d), material passing the #4 sieve (4.76 mm) and retained on a #16 sieve (1.19mm) was used.

2.1.2 Granite

In addition to the grout samples, a section of 10.2 cm (4") diameter core of Lac du Bonne granite was obtained for analysis. In order to obtain sample discs for the diffusion experiments of equivalent surface area as the grout material, a 5.08 cm (2") sub core was drilled using a laboratory diamond drill. Discs approximately 6 mm thick were cut using the diamond rock saw and then ground to a 4 mm thickness and processed as described above. Since the granite samples were not in contact with the SCSS solution prior to processing, they were stored in the desiccator, under vacuum, for one month before the diffusion tests were initiated. The excess granite material not required for the diffusion experiments was retained and processed for distribution coefficient tests in a manner similar to the grout material.

2.2 SCSS Solutions

A simulated saline groundwater, "Canadian Shield Saline Solution" (SCSS), was used in both the curing of the grout mixes and the diffusion and adsorption experiments. The chemical analysis of the initial SCSS solution used in the curing of the cement grout specimens is given in Table 2 along with an analysis of the solution after the curing period. As seen from Table 2, there is a significant change in the chemistries of the post and pre-curing solutions, particularly with respect to the Ca, Mg, K and Cl concentrations. The increase in pH derived from the highly alkaline nature of cement (portlandite and KOH solubility control) has caused the precipitation of Ca and Mg salts as evidenced by the dramatic decrease in the Cl, Ca, and Mg content of the post curing solution. In addition, the Na content and particularly the K content of the curing solution has increased as a result of the dissolution of KOH and NaOH from the cement cylinders.

In order to minimize the effect of further chemical reactions between the cement discs and the equilibrating solutions in the diffusion and adsorption experiments, the pH of the SCSS solution used in these experiments was raised from 5.95 to 12.5 by the addition of NaOH and then filtered through 0.1 μ m cellulose nitrate filters. The chemical composition of the SCSS solution used for the tests on the grout material is shown in Table 3. The SCSS solution used in the experiments conducted on the granite discs was not pH adjusted and its chemical composition is given in the first column of Table 3.

Table 3: Standard Canadian Shield Saline Solution
Chemical Analyses of Equilibrating Solutions

	Initial Solution Concentration (mg/L)	Equilibrating Solution* Concentration (mg/L)
Ca	15000	15000
Mg	185	0.09
Na	4900	6450
K	47	35
Sr	33	35
SO ₄	< 500	735
Cl	36000	37000
TIC	0.6	1.4
pH	5.95	12.5
* samples filtered through 0.1 μ m		

2.3 Distribution Coefficients

Distribution coefficients for strontium and cesium were determined by conventional equilibrium batch methods. In this procedure, 10 g of the 1.19 mm to 4.76 mm size fraction of the sample were placed in a sealable polypropylene container to which 100 mL of equilibrating solution were added. The equilibrating solution used was SCSS solution (as described above) to which either radioactive ^{85}Sr or ^{137}Cs was added to allow for radiometric analyses. One concentration of Sr (3.76×10^{-4} M for the granite tests and 4.00×10^{-4} M for the grout experiments) and three Cs concentrations (2.75×10^{-7} M, 1.83×10^{-7} M and 1.74×10^{-7} M) were used. It should be noted that approximately 1.74×10^{-7} M of Cs and 3.99×10^{-4} M of Sr were introduced as impurities into the SCSS solution, thus restricting the range of Cs and Sr concentrations that could be considered.

Blank samples containing only the spiked SCSS solution were included for use as standard values in order to account for the possibility of sorption on the container walls. The kinetics of adsorption were then determined by removing and counting a 1 or 2 mL aliquot of the supernatant over time. The ^{85}Sr or ^{137}Cs activity was expressed as counts per minute (cpm) and compared to the activity of an equal volume of the standard solution (spiked SCSS solution). The difference in activity was taken to be equal to the amount of tracer associated with the solid phase. All analyses were performed in triplicate.

Distribution coefficients (K_d) were obtained by mass balance according to:

$$K_d = \frac{C_{\text{std}} - C_{\text{sol}}}{C_{\text{sol}}} \times \frac{V}{M} \quad (1)$$

where

C_{std} = activity in the initial SCSS solution corrected for container sorption effects (cpm)

C_{sol} = activity in the solution phase in contact with the solid phase (cpm) at a specific time

V = volume of the solution added to the solid phase (mL)

M = oven-dry mass of the solid phase (g)

2.4 Diffusion Coefficients - Measurement and Calculation

In the diffusion experiments, a one-dimensional, closed-system diffusion cell was used. The basic cell consists of two 125 mL reservoirs separated by the sample disc; one reservoir contained the active SCSS solution (spiked with tritiated water (^3H) and ^{36}Cl) while the other reservoir (nonactive) contained the SCSS solution described in Table 3. A schematic of the diffusion cell is shown in Figure 1 and detailed information regarding its construction and the cell filling and sampling procedure is given in Johnston and Wilmot (1988).

To ensure a constant tracer concentration at the disc/active reservoir interface, the active SCSS solution was circulated from a central stock reservoir by a peristaltic pump at a rate of 0.65 to 0.75 mL/min. By using two peristaltic pumps, up to 20 cells could be accommodated, thus enabling several tests to be performed simultaneously. All diffusion experiments were conducted at room temperature ($\sim 21^\circ\text{C}$).

Assuming tritiated water and chloride to be conservative in nature, the effective diffusion coefficients for tritium and ^{36}Cl through the granite/grout discs were calculated from the mass transfer of the tracer, by diffusion, through the sample from time t_1 to t_2 , as illustrated in Figure 2. The flux (F) at the interface $x=l$ is described by Fick's first law:

$$F = -\eta D \left. \frac{dC}{dx} \right|_{x=l} \quad (2)$$

where D is the effective diffusion coefficient and includes the effect of tortuosity, η is the porosity, C is the concentration of the tracer of interest and x is a spatial coordinate. The flux is also given by the total mass of the solute in a volume V at time t per unit area of the disc (A):

$$F = \frac{V}{A} \frac{dC_t}{dt} \quad (3)$$

Figure 1: Schematic of the Diffusion Cell

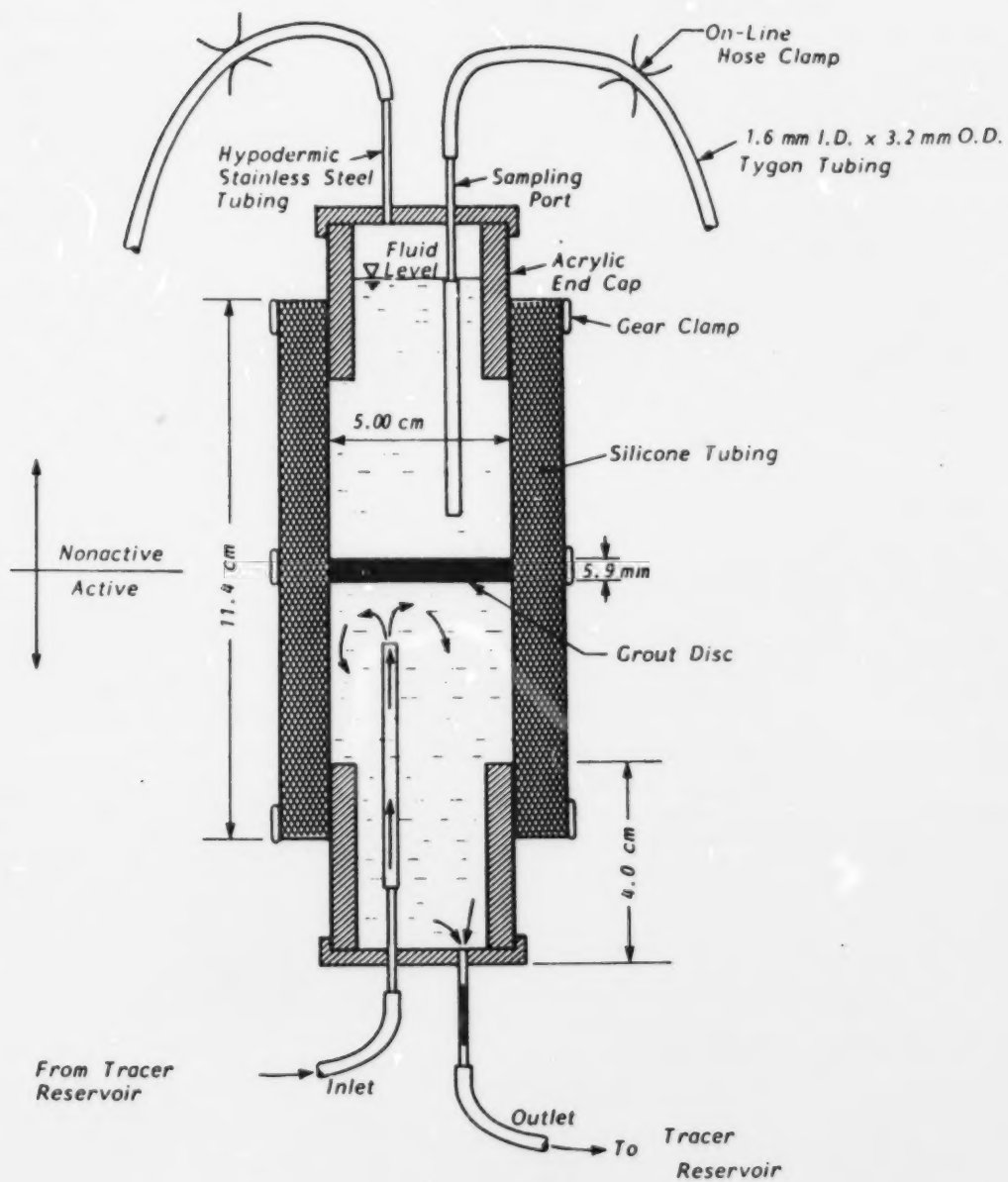
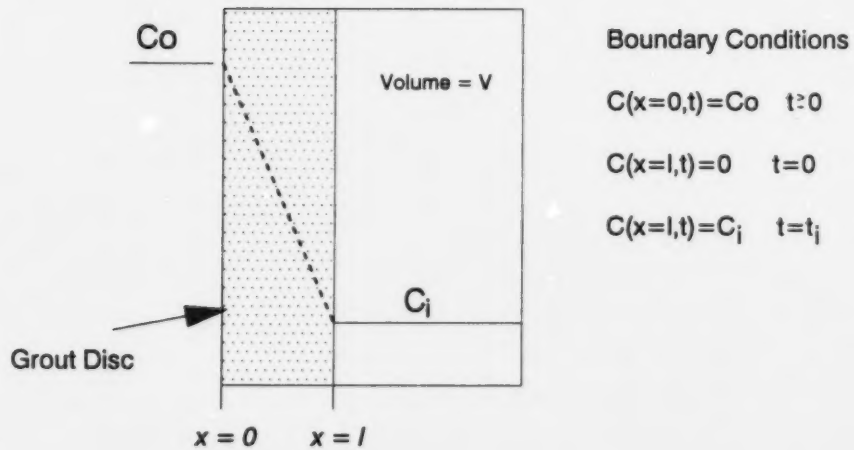


Figure 2: Illustration of the Diffusion Process



If the sample disc is relatively thin and we assume steady-state conditions and a homogeneous sample, then the concentration gradient within the granite/grout sample should be linear. In addition, if $C < C_0$ equations (2) and (3) can be simplified and combined to give:

$$\eta D_{(t_1-t_2)} = \frac{Vl}{AC_0} \frac{C_2 - C_1}{t_2 - t_1} \quad (4)$$

where

- D = effective diffusion coefficient measured between times t_1 and t_2 (L^2/T)
- η = porosity of the sample disc
- C_t = activity of tracer in the nonactive reservoir at time t (cpm or dpm)
- C_0 = activity of tracer in the active reservoir (cpm or dpm)
- V = volume of solution in the nonactive reservoir (L^3)
- A = surface area of the sample disc (L^2)
- l = disc thickness (L)
- t = time (T)

The observed diffusion coefficients were determined by least squares regression of $C_t V / C_0 A$ versus time (t) once linearity of the graph had been visually established, as illustrated in Figure 3.

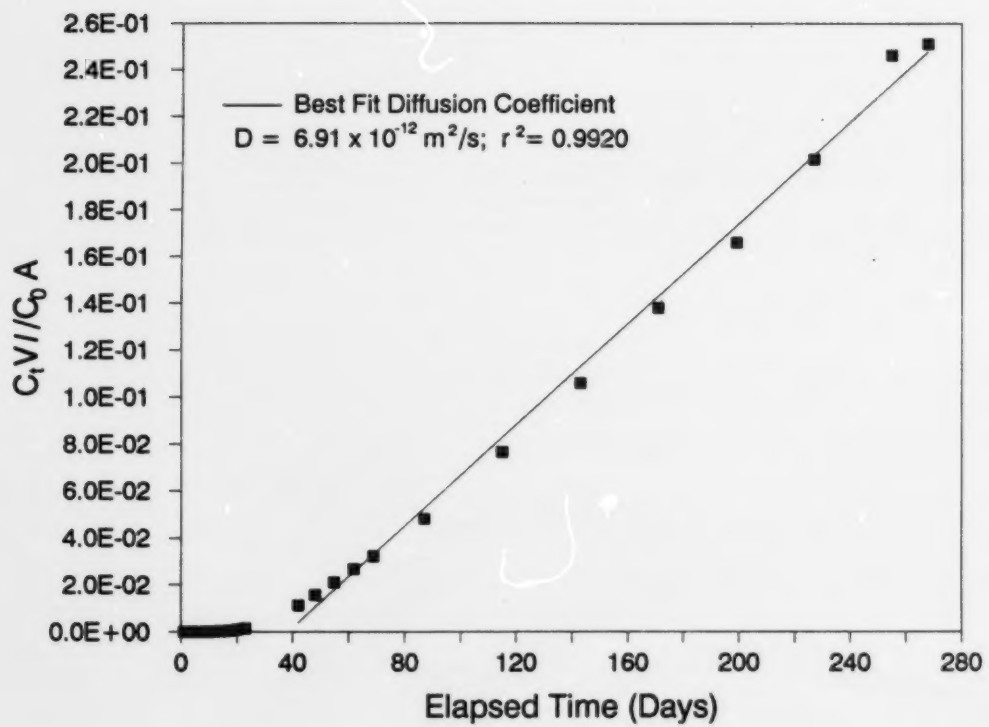
3.0 RESULTS AND DISCUSSION

3.1 Retardation Parameters

The distribution coefficient (K_d) is a partitioning parameter defined as the concentration of the solute in the solid phase divided by the concentration in the solution phase. It is applicable to describe uptake reactions that are instantaneous and reversible and in which there is a functional relationship between the solution and solid phase concentrations. If the functional relationship is linear then a retardation factor (R) can be defined by:

$$R = 1 + \left(\frac{\rho}{\eta}\right) K_d \quad (5)$$

Figure 3: Regression Results for ^{36}Cl Diffusion Data for
Grout Mix 100% T50, w/c=0.35, Replicate 1



where η is the effective porosity (L^3/L^3) and ρ is the bulk density (M/L^3). Further, the diffusive migration rate of a reactive solute in a porous medium is given by:

$$D_s = \frac{D}{R} = \frac{D_0 \tau}{1 + \left(\frac{\rho}{\eta}\right) K_d} \quad (6)$$

where D_0 is the free solution diffusion coefficient of the solute of interest and τ is the tortuosity factor of the medium. Other researchers have found that uptake of both Cs and Sr by cement-based materials can be adequately described by the use of a distribution coefficient (e.g. Atkinson and Nickerson, 1986) and thus use of a retardation factor in estimating diffusive migration rates for Cs and Sr is a reasonable approach.

The results of the equilibrium batch experiments for cesium and strontium are summarized in Table 4 and the raw data are listed in Appendix B. The results given in Table 4 are the mean and standard deviation of the constant point values on the linear portion of the measured distribution curves over the concentration ranges studied (values constant over time and equilibrium solution concentration; Appendix B).

3.1.1 Grout Sorption Results

An example of the equilibrium batch test results for the 85% T50:15% SF grout sample at a water to cement ratio of 0.35 is given in Figure 4. Generally, as seen from Figure 4, more scatter in the data was observed in the Sr uptake experiments relative to the Cs tests and this is reflected in the relatively higher standard deviation values for the Sr distribution coefficients presented in Table 4. Cesium adsorption coefficients ranged from 0.12 mL/g to 0.31 mL/g while Sr K_d values varied from 0.1 mL/g to 0.19 mL/g. The magnitude of these K_d values indicate that neither Cs nor Sr is absorbed very strongly for any of the cement mixes and that Sr was sorbed to a lesser extent than Cs. This is opposite to the general trend observed by other researchers (Atkinson and Nickerson, 1986; Plećaš et al, 1990) and is likely due to the high ionic strength of the SCSS solution.

Averages of the replicates values for the measured distribution coefficients are shown in Figure 5 as a function of the T50 cement content. The K_d values for Cs increase by a factor of 2 to 3 with the increasing percentage of silica fume. This trend is also observed for the Sr K_d data for grout mixes having a w/c ratio of 0.25. Also evident from Figure 5 is the increase in K_d values with decreasing water to cement ratios. This trend is anticipated due to the higher surface areas available for sorption due to the increasing

**Table 4: Summary of Strontium and Cesium
Distribution Coefficient Results**

Mix	Water to Cement Ratio	Replicate	Distribution Coefficient (mL/g)	
			Sr	Cs
100% T50	0.35	1	0.178 (.065) *	0.110 (.043)
		2	0.189 (.066)	0.124 (.053)
		3	0.178 (.062)	0.123 (.053)
	0.25	1	0.118 (.070)	0.117 (.043)
		2	0.088 (.062)	0.121 (.059)
		3	0.098 (.067)	0.117 (.047)
90% T50 10% SF	0.35	1	0.099 (.071)	0.149 (.046)
		2	0.124 (.069)	0.147 (.050)
		3	0.091 (.052)	0.161 (.047)
	0.25	1	0.142 (.077)	0.206 (.046)
		2	0.137 (.077)	0.200 (.037)
		3	0.133 (.061)	0.207 (.052)
85% T50 15% SF	0.35	1	0.200 (.073)	0.230 (.052)
		2	0.184 (.058)	0.256 (.044)
		3	0.171 (.062)	0.252 (.046)
	0.25	1	0.173 (.069)	0.310 (.056)
		2	0.173 (.083)	0.305 (.060)
		3	0.176 (.072)	0.307 (.051)
Granite	n/a	1	0.017 (.021)	3.46 (.24)
		2	0.021 (.030)	3.27 (.21)
		3	0.029 (.023)	3.25 (.25)
*(standard deviation) n/a - not applicable				

Figure 4: Cs and Sr Kd Results for Grout Mix 85% T50-15% SF
(w/c=0.35 in SCSS)

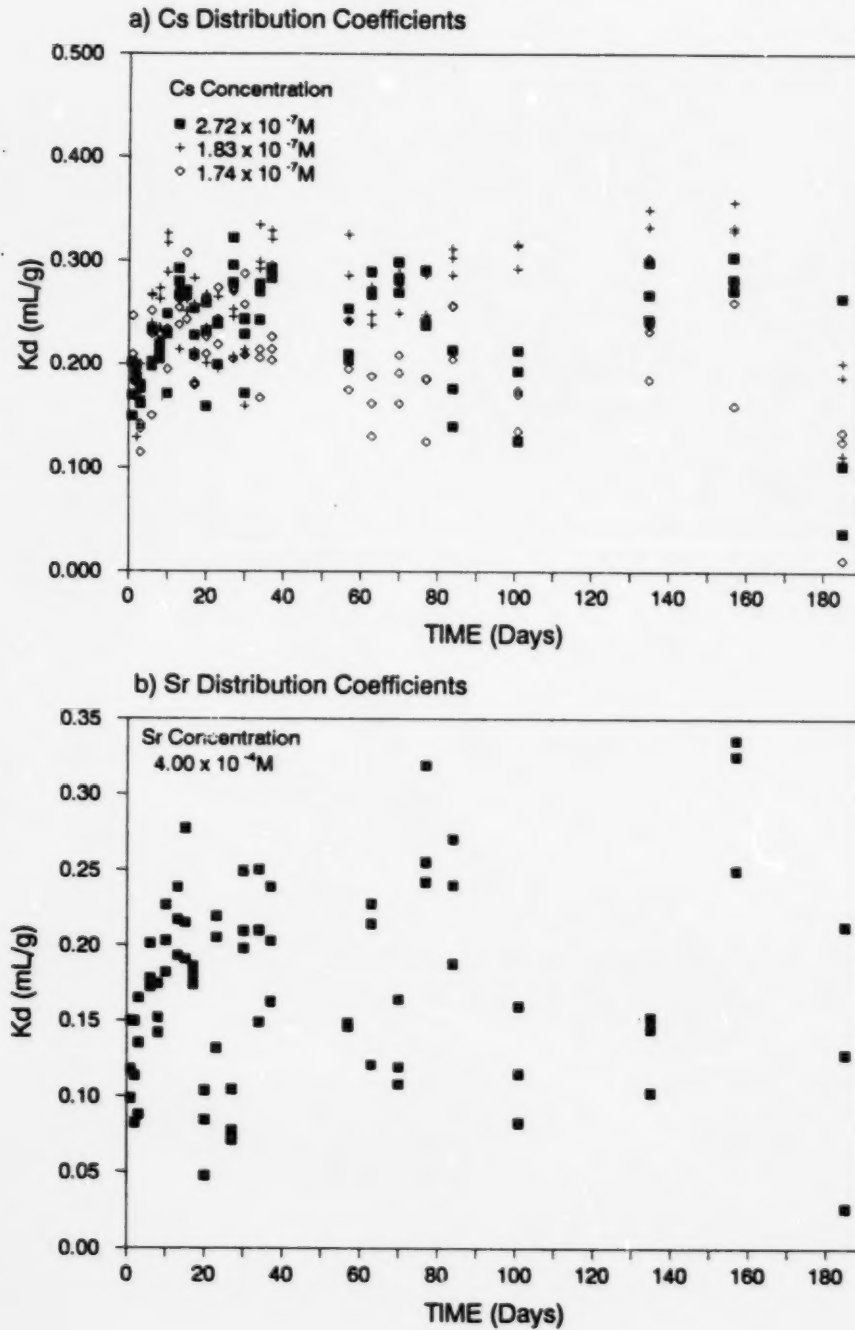
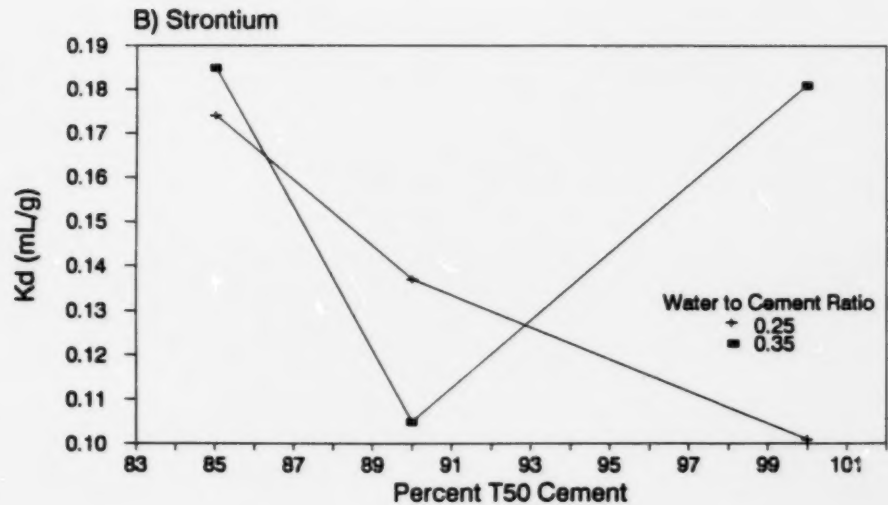
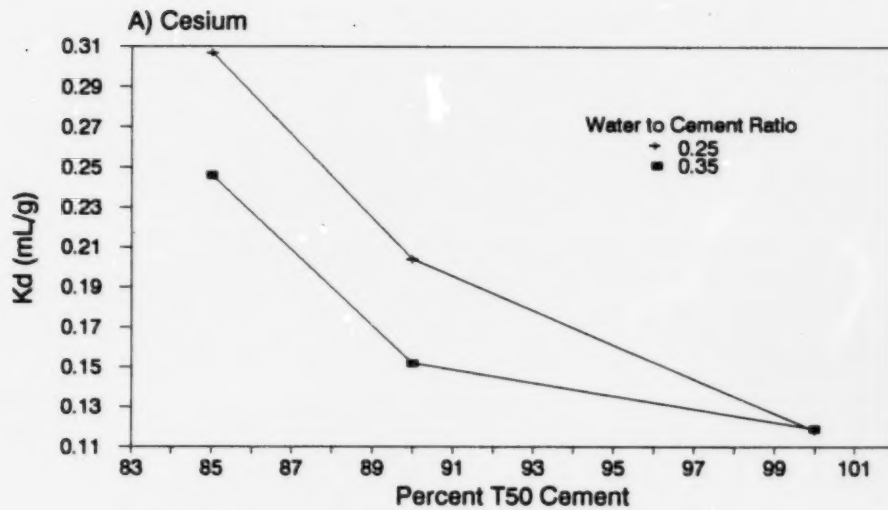


Figure 5: Summary of Cs and Sr Distribution Coefficient Results
for Cement Grout Mixes in SCSS



silica fume content and decreasing water to cement ratios.

The porosity and bulk density values measured for the grout samples were given in Table 1. As one would expect, the porosity values decrease as the water to cement ratio decreases for each of the grout mixes, however, the porosity of the 100% cement samples are consistently lower than those for the silica fume mixes at the corresponding water to cement ratio. The bulk densities, on the other hand, exhibit a different trend with ρ decreasing as the percent of silica fume increases due to the lower specific gravity of silica fume relative to that of unreacted cement.

Calculated retardation factors for both Cs and Sr are presented in Table 4 and are shown graphically in Figure 6. It is interesting to note that there is no significant difference shown between the retardation factors calculated for the 100% T50 grout samples and the 90% T50:10% SF samples even though the measured distribution coefficients for both Cs and Sr increased with the addition of silica fume. Figure 7 illustrates the reason for this behaviour. Equation (6) shows that the retardation factor is proportional to the distribution coefficient and the bulk density but is inversely proportional to the porosity. As seen in Figure 6, although the Cs Kd value increases from 0.12 mL/g to 0.31 mL/g as the percent of silica fume increases from 0% to 15% (w/c=0.25), the corresponding bulk density to porosity ratios decrease from 0.35 to 0.22. For the 100% T50 cement and the 90% T50:10% SF samples, the almost three-fold increase in Kd was offset by the decrease in the bulk density to porosity ratio. These results show that the variations in bulk density and porosity can have a significant effect on the retardation factor and thus the migration rate of reactive solutes in grout materials and is similar to the results observed by Gillham et al (1989) for diffusion of Sr in bentonite-sand mixtures. Indeed, Figure 7 indicates that, with respect to the migration rates of ^{85}Sr or ^{137}Cs in a saline groundwater environment, an excess of 10% silica fume would be required in order to significantly increase the retardation capability of the cement-based grout.

3.1.2 Granite Sorption Results

The Kd results of the Cs sorption experiments conducted on samples of the Lac du Bonne batholith using SCSS are shown graphically in Figure 8. Generally, the plots of the distribution coefficient versus time exhibit the classic form of the distribution curve, i.e. an initial period of increasing Kd until ion-exchange equilibria is attained after which a relatively constant distribution coefficient is observed. As seen from Figure 8, little scatter was observed in among the replicate samples. The results presented in Table 4 are the combined values of the horizontal portion of the distribution curves for both the 1.04×10^{-8} M and 5.89×10^{-10} M concentrations giving average Kd values of 3.46 ± 0.24 mL/g, 3.27 ± 0.21 mL/g and 3.27 ± 0.25 mL/g for sample replicates 1, 2 and 3 respectively. These

Figure 6: Calculated Cs and Sr Retardation Factors

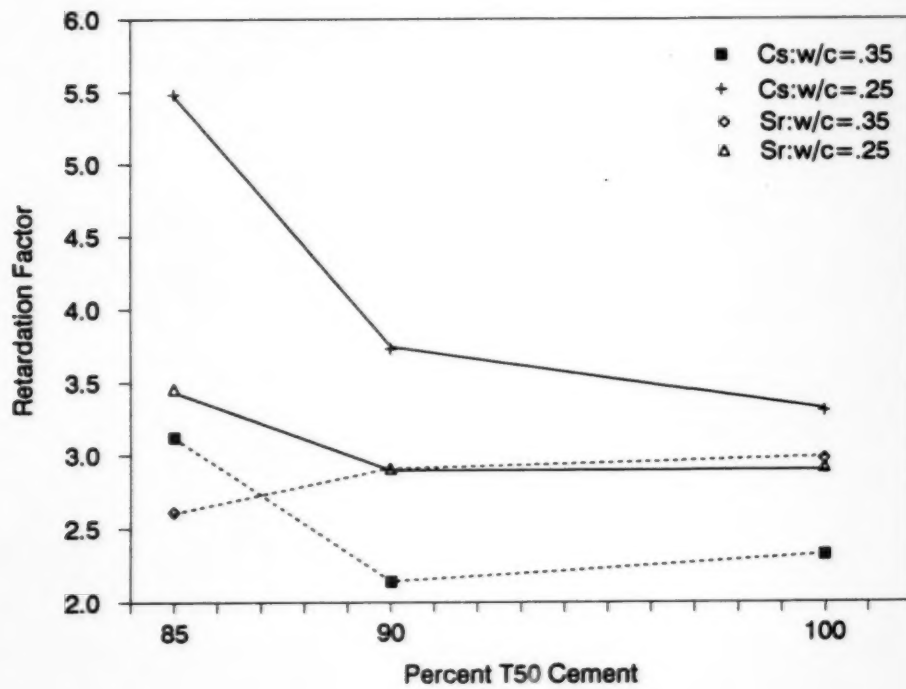


Figure 7: Cesium Kd and Bulk Density/Porosity Ratios
vs %T50 Cement

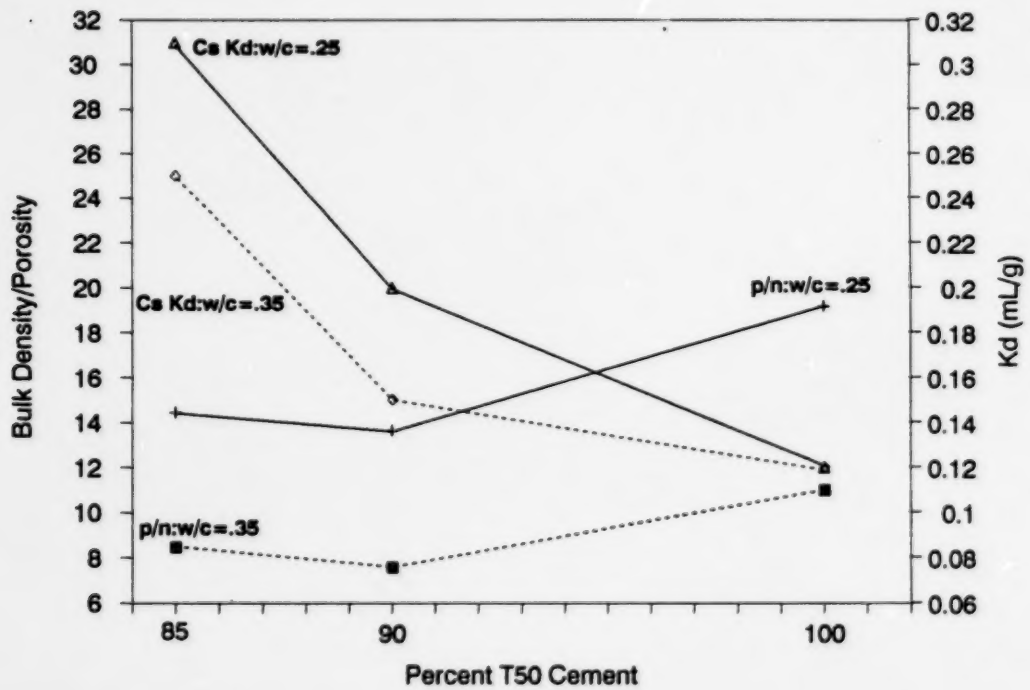
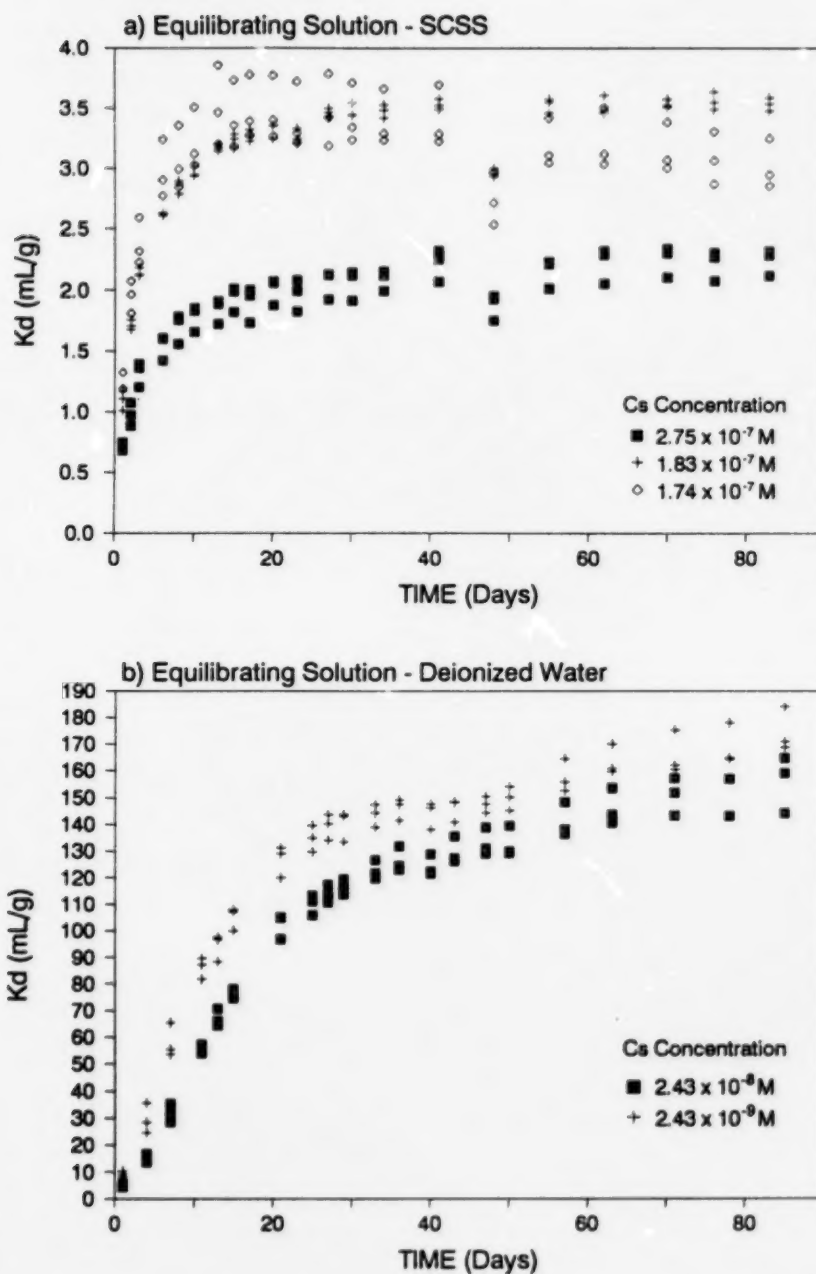


Figure 8: Granite Rock Core Cs Kd Results



values are over an order of magnitude lower than the Cs distribution coefficients determined on granite samples using deionized water as the equilibrating solution (Figure 8b). In the deionized water tests, two Cs concentrations were considered, 2.43×10^{-8} M and 2.43×10^{-9} M, giving Cs Kd values of 162.3 ± 10.1 mL/g and 174.8 ± 7.5 mL/g, respectively. These deionized Kd values compare well with the results obtained using samples of Precambrian granitic gneiss for which a distribution coefficient for Cs of $\sim 88 \pm 10$ mL/g was obtained (Johnston and Wilmot, 1988). The significant reduction in Kd values measured for Cs (and also for Sr) using SCSS is due to the high ionic strength of the solution (~ 1.43 M).

Equilibrium batch test results for Sr are shown on Figure 9 and summarized in Table 4. As seen from Figure 9, the Sr Kd values are very low (0.022 ± 0.026 mL/g) and the data points exhibit a large degree of scatter. As was observed for the Cs results, distribution coefficients determined for Sr using deionized water were much higher, on the order of 4.0 ± 0.4 mL/g, and the data showed much less variability about the mean. Again the deionized Sr Kd values obtained for the sample of Lac du Bonne granite were comparable to that measured for the Precambrian granitic gneiss (2.93 ± 0.43 mL/g; Johnston and Wilmot, 1988).

3.2 Effective Diffusion Coefficients

The experimental diffusion profiles for ^3H and ^{36}Cl are for the cement grout mixes and Lac du Bonne granite are shown both in tabular and graphic form in Appendix C. As stated in section 2.4, diffusion coefficients were obtained by linear regression of the C_i/C_0 versus time data once linearity had been visually established. The regression equations and number of data points used in the regression process is given in Appendix B and is graphically represented by the length of the fitted diffusion curves depicted in the graphs. For all of the samples studied the measured concentration data shows good agreement with calculated C_i/C_0 values based on Fick's first law of diffusion (equation 4).

3.2.1 Grout

A summary of the results of the diffusion experiments for the cement grouts are shown in Table 5 and examples of the ^3H and ^{36}Cl concentration versus time profiles obtained for the 85% T50 cement:10% SF grout ($w/c=.25$) are shown in Figures 10 and 11, respectively. The measured results were found to be reproducible and the degree of scatter of data points about the best-fit curves was quite small, as evidenced by r^2 values equal to 0.9961 and 0.9927 for the tritium replicates and 0.9953 and 0.9936 for the chloride diffusion results (see Figures 10 and 11). The results of the diffusion experiments

Figure 9: Granite Rock Core Sr Kd Results

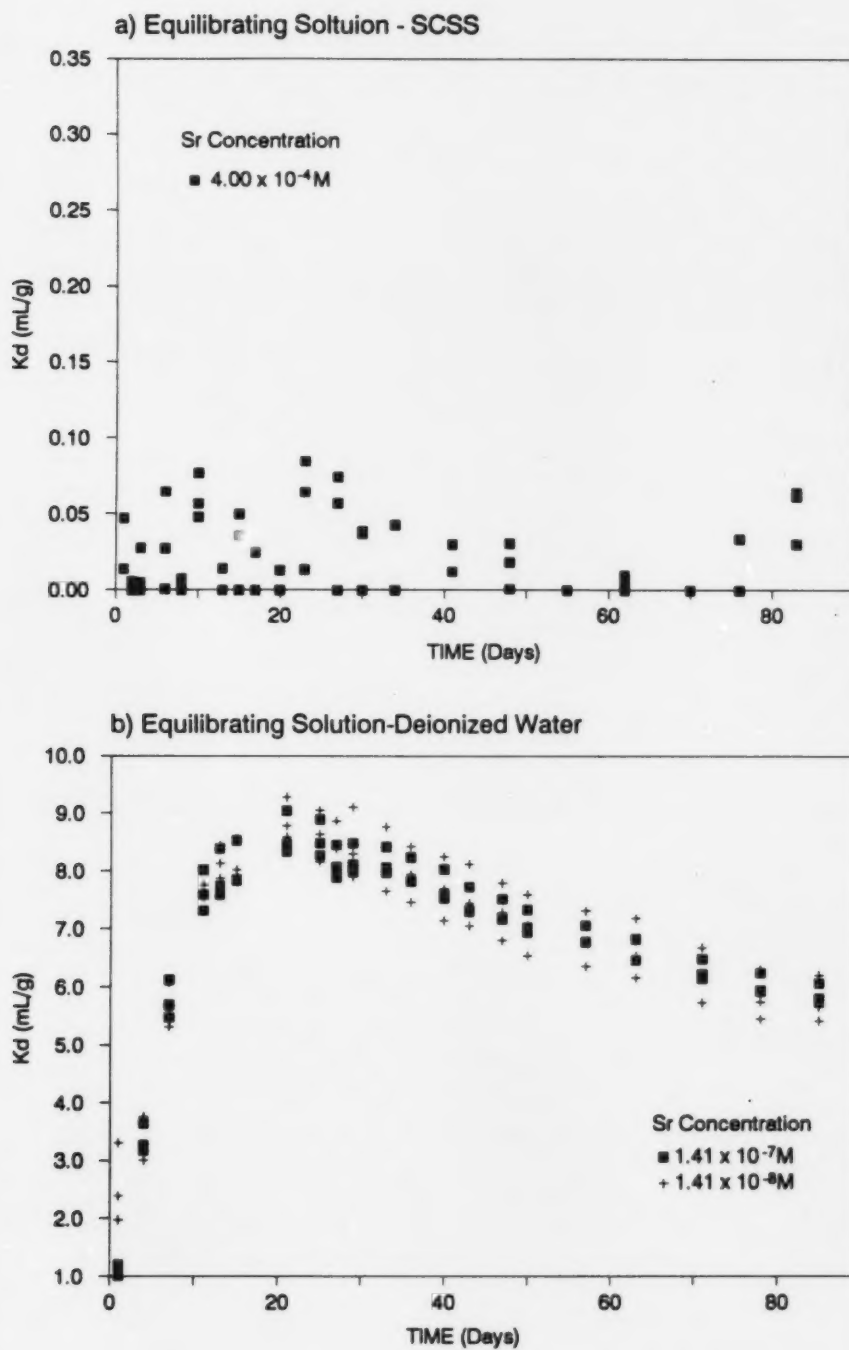


Figure 10: Tritium Diffusion Results for Grout Mix
85% T50:15% SF (w/c=0.25 in SCSS)

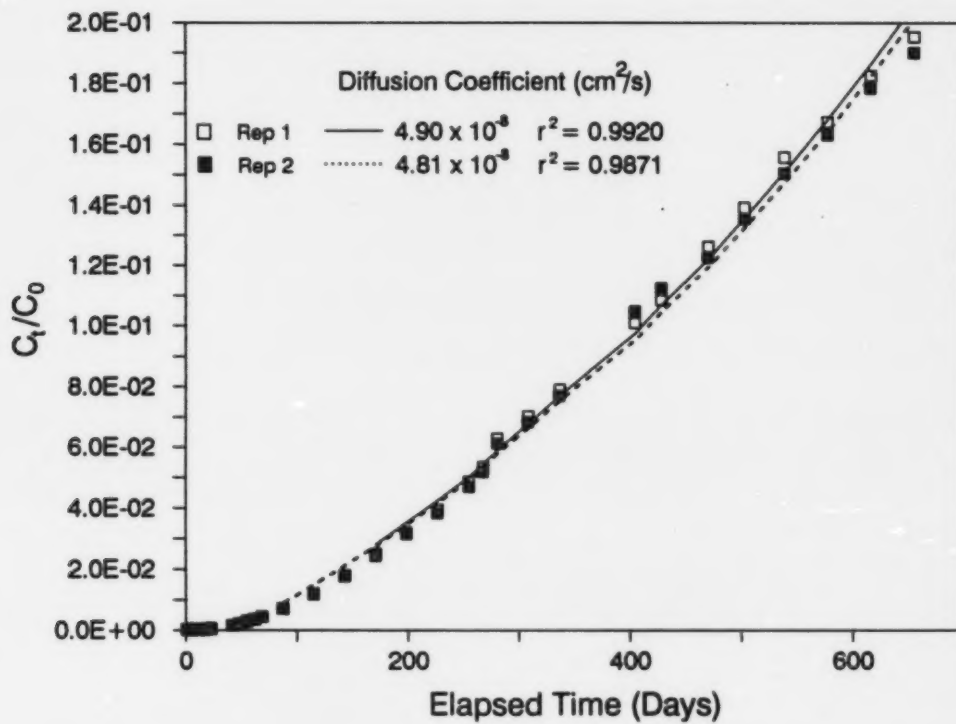
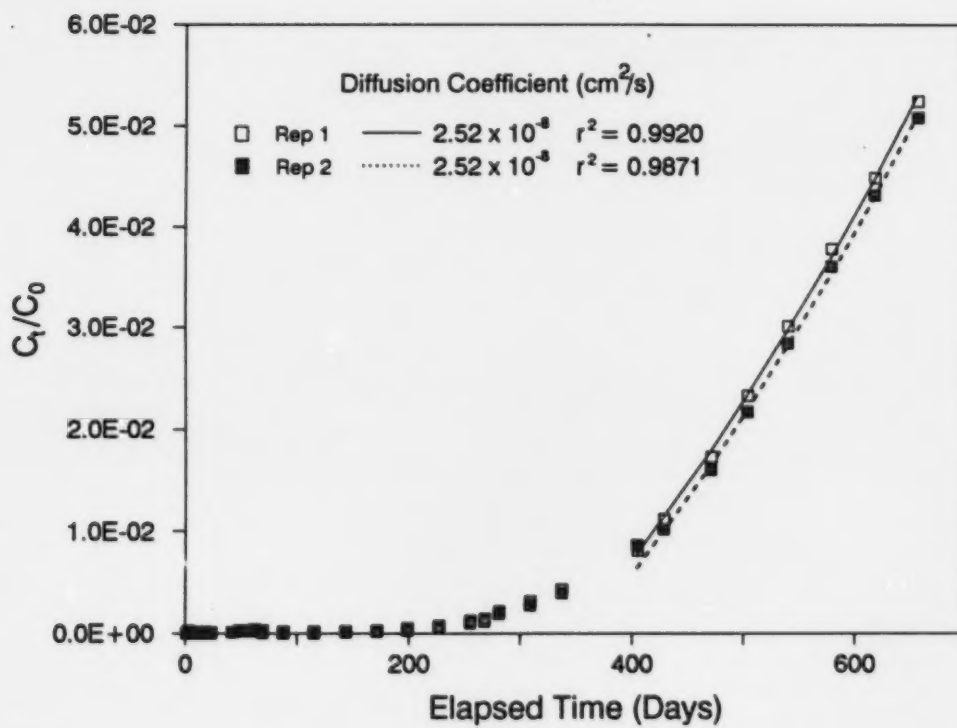


Figure 11: Chlorine-36 Diffusion Results for Grout Mix
85% T50:15% SF (w/c=0.25 in SCSS)



also show, as is evident from Figure 11, that extremely long lag times (eg. ~350 days for ^{36}Cl) were observed for several of the grout mixes before the diffusive flux reached steady state. This was particularly true of the chloride diffusion test results with lag time increasing with the amount of silica fume in the mix and with decreasing water to cement ratios.

The time to reach steady-state in the diffusion experiments is a function of reaction processes. For example, if ion-exchange is the principal reaction mechanism then the observed time lag (t_o) can be related to the retardation factor (R) by:

$$R = \frac{6 D t_o}{l^2} \quad (7)$$

where

- D = the diffusion coefficient (L^2/T)
- t_o = the time lag (T)
- l = the disc thickness (L)

In relation to a distribution coefficient (Kd), equation (7) can be re-written as:

$$Kd = \left(\frac{6 D t_o}{l^2} - 1 \right) \frac{\eta}{\rho} \quad (8)$$

A comparison of predicted time lag (assuming no sorption) to the observed time lag shows that the approach to steady state (equilibrium within the cement slab) was, in most cases, much slower than expected. For example, if we assume no sorption or reaction occurs between the radioactive tracer and the grout, then substitution of the measured diffusion coefficient at steady-state for the 85% T50:15% SF mix at a w/c=0.25 and a Kd=0 into equation (8) gives an expected time lag of ~13 days for ^3H and ~27 days for ^{36}Cl . However, the actual observed time lag, as seen in Figures 10 and 11, was ~44 days and ~350 days for tritium and ^{36}Cl , respectively. As a general trend, the measured time lag increased with the amount of silica fume by a factor of 2-4 for tritium and by a factor of 10 for chloride. With respect to ^3H , isotopic exchange with water molecules associated with cementitious mineral phases is a likely explanation, however this was not investigated further. Chloride, on the other hand, has been shown to undergo substantial interactions with C-S-H (calcium hydrosilicate) phases in the cement (Beaudoin et al, 1990; Tritthart, 1989). For example, Tritthart (1989) investigated the chemical composition

of cement porewaters in relation to the initial curing water composition. His findings showed that for ordinary portland cement ($w/c=0.5$) cured for 28 days in a 3000 ppm Cl solution, the percent Cl in the cement mass at 28 days was 0.35% and this increased to ~0.50% after 112 days. In fact, it took over 200 days before the Cl content in the porewater reached equilibrium with the curing water. Since our samples were cured for 58 days prior to use in the diffusion tests it is conceivable that a portion of the ^{36}Cl lost from the solution phase during the diffusion experiments is a result of incomplete chloride binding (disequilibrium) prior to the initiation of the diffusion experiments. In addition to this factor, there would also be uptake of ^{36}Cl as a result of isotopic exchange similar to that postulated for tritium. The time lag results indicate that with respect to cement-based materials, neither tritium nor chloride can be considered as unreactive tracers and that, considering the magnitude of the time lag with respect to ^{36}Cl , the degree of chloride binding in cement mixes increases significantly with percent silica fume.

The measured diffusion coefficients obtained for the samples of cement grout varied from $1.12 \times 10^{-7} \text{ cm}^2/\text{s}$ to $4.90 \times 10^{-8} \text{ cm}^2/\text{s}$ for tritium and from $7.46 \times 10^{-8} \text{ cm}^2/\text{s}$ to $2.52 \times 10^{-8} \text{ cm}^2/\text{s}$ for ^{36}Cl depending on the mix and water to cement ratio used. These values are in general agreement with other reported results (Atabek et al, 1990; Diab et al, 1988, Kumar, 1985; Byfors, 1987). The effect of silica fume and water to cement ratio on tritium diffusion coefficients is shown in Figure 12. The measured ^3H diffusion coefficients decrease by approximately half an order of magnitude from an average value of $8.45 \times 10^{-8} \text{ cm}^2/\text{s}$ for the 100% T50 grout to $4.65 \times 10^{-8} \text{ cm}^2/\text{s}$ for the 85% T50:15% SF grout mixed at a $w/c=0.35$. Similar trends were observed for the $w/c=0.25$ grout mixes for both the tritium and ^{36}Cl tracers. In addition, decreasing the water to cement ratio from 0.35 to 0.25 resulted in a further reduction in the magnitude of the measured diffusion coefficient. Figure 12 also indicates that, with respect to limiting the mobility of radioactive contaminants, reducing the water to solids ratio of the 90% T50 cement:10% silica fume from 0.35 to 0.25 would be equivalent to adding 15% silica fume to the mix but maintaining a higher water to cement ratio.

3.2.2 Granite

The experimental diffusion profiles for ^3H and ^{36}Cl are shown in tabular form in Table 5 and graphically in Figure 13. As seen in Figure 13, the data were reproducible, exhibiting remarkable little scatter in the C/C_0 values with respect to the fitted curves (r^2 values ranged from 0.9824 to 0.9989). The measured diffusion coefficients obtained for samples of the Lac du Bonne granite ranged from $1.51 \times 10^{-6} \text{ cm}^2/\text{s}$ to $1.57 \times 10^{-6} \text{ cm}^2/\text{s}$ for tritium and $9.6 \times 10^{-7} \text{ cm}^2/\text{s}$ to $1.01 \times 10^{-6} \text{ cm}^2/\text{s}$ for ^{36}Cl . These values compare well with those reported in the literature. For example, earlier work conducted on Precambrian granite samples using deionized water gave diffusion coefficients on the order of $2.6 \times 10^{-7} \text{ cm}^2/\text{s}$

Table 5: Summary of Tritium and Chloride
Diffusion Results Using SCSS

Mix	Water to Cement Ratio	Replicate	D x10 ⁸ (cm ² /s)		Tortuosity Factor* (τx10 ³)	
			³ H	³⁶ Cl	³ H	³⁶ Cl
100% T50	0.35	1	8.18	6.91	1.73	3.80
		2	8.67	7.46	1.83	4.09
	0.25	1	11.24	8.84	2.37	3.76
		2	7.01	3.51	1.48	1.93
90% T50 10% SF	0.35	1	5.20	3.04	1.10	1.67
		2	5.38	3.70	1.14	2.03
	0.25	1	5.77	1.41	1.22	0.78
		2	6.06	1.64	1.32	0.90
85% T50 15% SF	0.35	1	4.65	1.65	0.95	0.90
		2	4.64	1.52	0.98	0.83
	0.25	1	4.90	2.52	1.04	1.39
		2	4.81	2.52	1.02	1.39
Granite	n/a	1	156.8	101.3	189.3**	55.6
		2	150.8	96.0	182.0**	52.8
		3	152.0	98.8	183.5**	54.3
Free Solution Diffusion Coefficients at 21 °C						
* Do ^{Cl} = 1.82 x 10 ⁻⁵ cm ² /s						
Do ^{OH} = 4.73 x 10 ⁻⁵ cm ² /s						
** Do ^H = 8.28 x 10 ⁻⁵ cm ² /s						

Figure 12: Effect of Silica Fume on ^3H Diffusion Coefficients

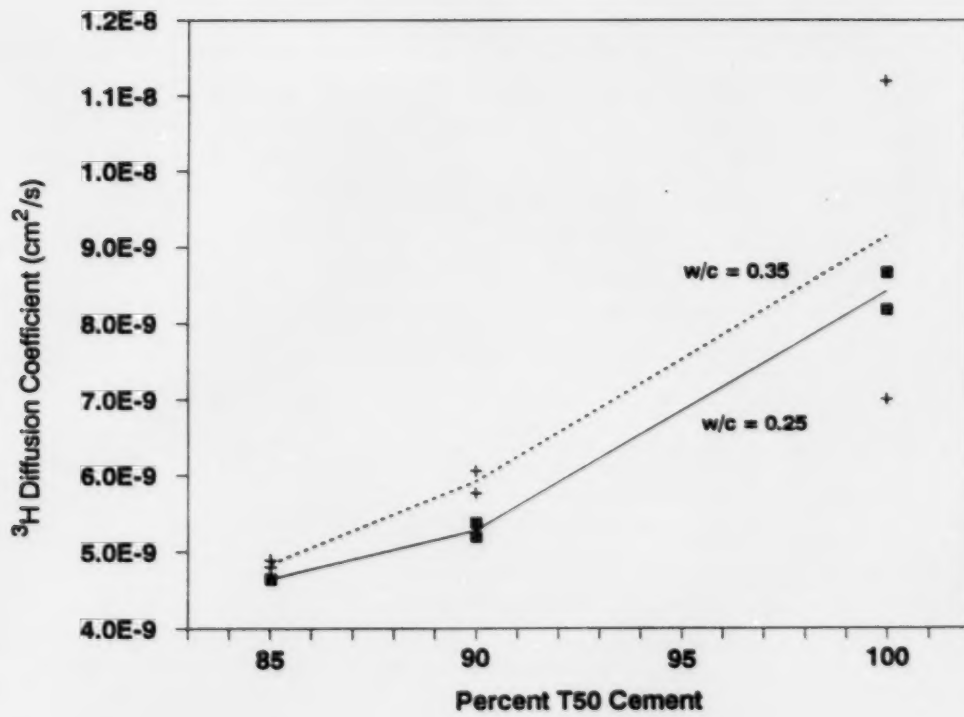
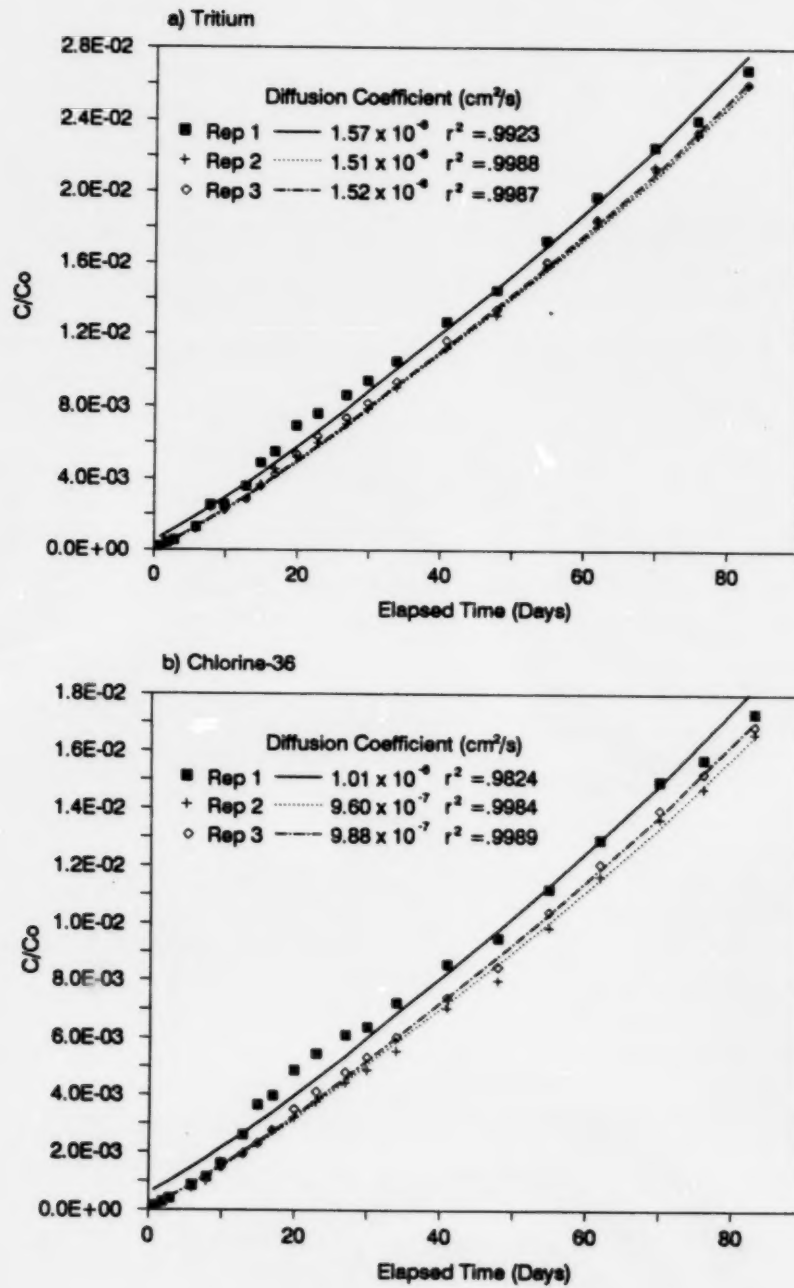


Figure 13: Tritium and ^{36}Cl Diffusion Results
for Lac du Bonne Granite



for ^3H and 1.3×10^{-7} to $2.9 \times 10^{-7} \text{ cm}^2/\text{s}$ for ^{36}Cl (Johnston and Wilmot, 1988). Bradbury et al (1982), in measuring diffusion coefficients on rock samples from four granitic formations in the United Kingdom, report iodide (I^-) diffusion coefficients in the range of 1.4×10^{-8} to $6.4 \times 10^{-9} \text{ cm}^2/\text{s}$. (It should be noted that these values include the effect of porosity and, as defined in this report, would be equivalent to ηD .) Although I^- was used as a tracer rather than chloride, because of the similarity in free-solution diffusion coefficients ($2.00 \times 10^{-5} \text{ cm}^2/\text{s}$ for I^- and $2.03 \times 10^{-5} \text{ cm}^2/\text{s}$ at 25°C) and anionic nature, one could anticipate comparable experimental results. Skagius and Neretnieks (1982) measure I^- diffusion coefficients on the order of 8.1 to $9.2 \times 10^{-7} \text{ cm}^2/\text{s}$ for samples of Finnsjö granite and 3.5×10^{-8} to $3.8 \times 10^{-7} \text{ cm}^2/\text{s}$ for Stripa granite while values of 1.4 to $2.0 \times 10^{-7} \text{ cm}^2/\text{s}$ were measured for tritium on samples of the Finnsjö granite. Subsequent work (Skagius and Neretnieks, 1986) on samples of Gideå gneiss resulted in I^- diffusion coefficient in the range of 8.3×10^{-7} to $1.1 \times 10^{-6} \text{ cm}^2/\text{s}$.

3.3 Calculated Cesium-137 and Strontium-85 Diffusion Coefficients for Cement Grouts

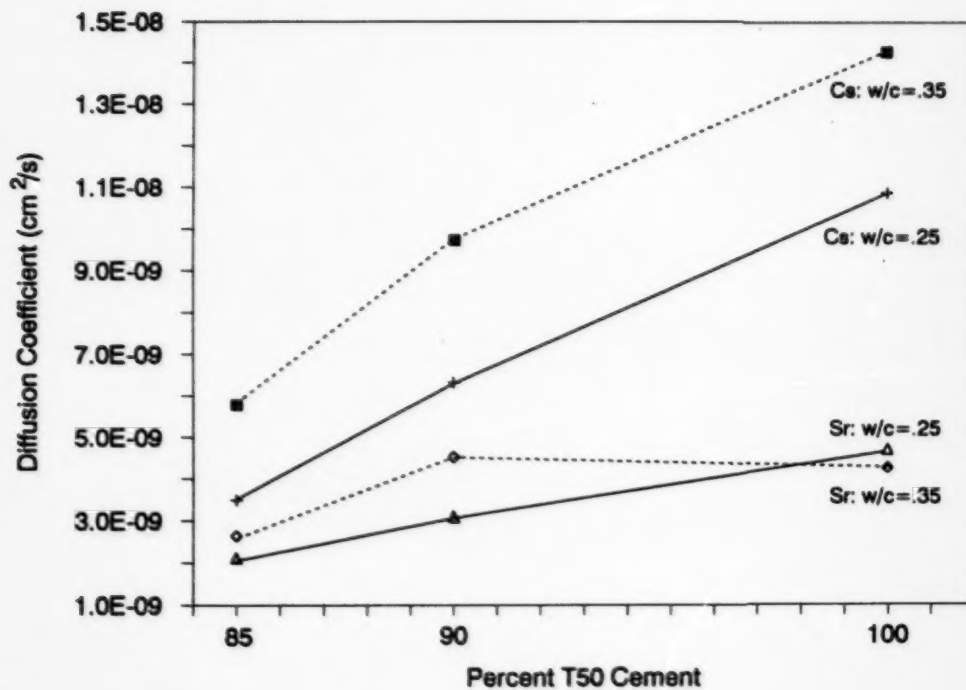
In order to estimate the effective diffusion coefficients of Cs and Sr in the candidate grout mixes, tortuosity factors were calculated using equation (6). Since a steady-state diffusion system was used the diffusion coefficients measured are independent of the reaction term. That is, all reaction sites on the grout samples would be occupied and thus $R=1$ (i.e. $K_d=0$). Therefore, the measured diffusion coefficient (D) would be equivalent to D_0 . The values of D_0 used were $1.82 \times 10^{-5} \text{ cm}^2/\text{s}$ for $^{36}\text{Cl}^-$ and $4.73 \times 10^{-5} \text{ cm}^2/\text{s}$ for O^{3-}H^- (Li and Gregory, 1974) corrected to 21°C . The calculated tortuosity factors are presented in Table 5. The tortuosity factors calculated from the ^{36}Cl diffusion results are in fairly good agreement with those obtained from the tritium diffusion coefficients, particularly for the higher silica fume grouts. Since there were generally more data points used in the regression analysis for the tritium diffusion results due to long time lags associated with the ^{36}Cl tests, the tortuosity parameters calculated from the ^3H diffusion experiments were used for the estimation of Cs and Sr diffusion coefficients.

An average value of the replicate values of the tortuosity factors obtained from the ^3H diffusion experiments were used in equation (6) to calculate ^{85}Sr and ^{137}Cs diffusion coefficients. The other parameters used in equation (6) were the measured porosity and bulk density of the grout mixes (Table 1), the average K_d values obtained from the equilibrium batch tests (Table 4), and values of $1.86 \times 10^{-5} \text{ cm}^2/\text{s}$ and $7.12 \times 10^{-6} \text{ cm}^2/\text{s}$ for the free solution diffusion coefficients (21°C) for cesium and strontium, respectively. The resulting ^{137}Cs and ^{85}Sr diffusion coefficients are shown in Figure 14 and presented in Table 6 along with the corresponding retardation factors and tortuosity factors.

Table 6: Calculated Cs and Sr Diffusion Coefficients
for the Cement Grout Mixes

Mix	Water to Cement Ratio	Tortuosity* ($\tau \times 10^3$)	Retardation Factor		Calculated Diffusion Coefficient** (cm^2/s)	
			Cs	Sr	Cs	Sr
100% T50	0.35	1.78	2.3	3.0	1.4×10^{-9}	4.2×10^{-9}
	0.25	1.93	3.3	2.9	1.1×10^{-9}	4.7×10^{-9}
90% T50 10% SF	0.35	1.12	2.2	1.8	9.5×10^{-9}	4.4×10^{-9}
	0.25	1.27	3.9	2.9	6.1×10^{-9}	3.1×10^{-9}
85% T50 15% SF	0.35	0.97	3.1	2.6	5.8×10^{-9}	2.7×10^{-9}
	0.25	1.03	5.4	3.5	3.5×10^{-9}	2.1×10^{-9}
* from tritium diffusion results ** using $\text{Do}^{\text{Ca}} = 1.86 \times 10^{-5} \text{ cm}^2/\text{s}$ $\text{Do}^{\text{Sr}} = 7.12 \times 10^{-6} \text{ cm}^2/\text{s}$						

Figure 14: Calculated Cs and Sr Diffusion Coefficients
for Cement Grouts



From Figure 14 it can be seen that, with respect to the calculated diffusion coefficients for ^{137}Cs and ^{85}Sr , the water to cement ratio has a significant effect on the estimated D values with both isotopes being less mobile at the lower water to cement ratios. It is interesting to note, however, that the amount of silica fume present has a much less pronounced effect on the effective diffusion coefficients. In fact, except for the case of ^{137}Cs diffusion in the 85% T50:15% SF grout at a w/c=0.35, an increase in silica fume from 0% to 15% had very little effect at all. Cesium diffusion coefficients at a w/c ratio of 0.25 decreased from $1.1 \times 10^{-8} \text{ cm}^2/\text{s}$ to $3.5 \times 10^{-9} \text{ cm}^2/\text{s}$ while ^{85}Sr diffusion coefficients were generally reduced by less than a factor of two ($4.7 \times 10^{-9} \text{ cm}^2/\text{s}$ to $2.1 \times 10^{-9} \text{ cm}^2/\text{s}$). It should be noted that these calculated diffusion coefficients are subject to the uncertainties associated with the measured ^3H diffusion coefficients (through the τ values) and calculated retardation factors (Kd's). Nevertheless, the apparent trend is significant in that, for the conditions studied, although the addition of silica fume to the cement is beneficial, from a contaminant transport point of view, a similar impact on the diffusive migration rates can be achieved through the use of lower water to cement ratios.

4.0 SUMMARY AND CONCLUSIONS

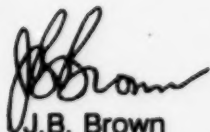
- 1.0 Cesium distribution coefficients for the cement grout ranged from 0.12 mL/g to 0.31 mL/g while Sr Kd values varied from 0.1 mL/g to 0.19 mL/g. The magnitude of the Kd values indicate that neither Cs nor Sr is adsorbed very strongly for any of the cement grout mixes due to the high ionic strength of the SCSS solution (~1.43 M).
- 2.0 The adsorption results indicate that a reduction in the water to cement ratio of a grout material was more effective in retarding the migration of ^{137}Cs and ^{85}Sr than was the addition of silica fume. Under the experimental conditions of this study, although the distribution coefficients were observed to increase as the percent silica fume increased from 0% to 15%, the calculated retardation factors did not exhibit the same trend due to a corresponding decrease in the bulk density to porosity ratio. This suggests that in a saline groundwater environment, an excess of 10% silica fume was required to significantly increase the ^{137}Cs and ^{85}Sr retardation capability of cement-based grout.
- 3.0 Batch test results using SCSS solution conducted on samples of the Lac du Bonne batholith gave Cs Kd values of ~3.3 mL/g and Sr Kd results of ~0.02 mL/g. These results are significantly lower than those obtained from similar tests conducted using deionized water (~175 mL/g and ~4.0 mL/g for Cs and Sr Kd's, respectively).

- 4.0 The measured diffusion coefficients obtained for the samples of cement grout varied from $1.12 \times 10^{-7} \text{ cm}^2/\text{s}$ to $5.20 \times 10^{-8} \text{ cm}^2/\text{s}$ for tritium, and from $7.46 \times 10^{-8} \text{ cm}^2/\text{s}$ to $1.41 \times 10^{-8} \text{ cm}^2/\text{s}$ for ^{36}Cl depending on the mix and water to cement ratio used.
- 5.0 In addition, results of the ^{36}Cl and tritium diffusion experiments showed that the addition of silica fume and the reduction of the water to solids ratio of the grout from 0.35 to 0.25 were advantageous with respect to minimizing diffusive migration.
- 6.0 The experimental diffusion profiles for ^3H and ^{36}Cl for samples of the Lac du Bonne granite ranged from $1.50 \times 10^{-8} \text{ cm}^2/\text{s}$ to $1.56 \times 10^{-8} \text{ cm}^2/\text{s}$ and from 9.6×10^{-7} to $1.01 \times 10^{-6} \text{ cm}^2/\text{s}$, respectively, and are of similar magnitude to diffusion coefficients for granitic rock reported in the literature.
- 7.0 Estimated effective diffusion coefficients for ^{137}Cs and ^{85}Sr also indicate that the water to cement ratio has a significant effect on their migration potential in cement-based grout with both radionuclides being less mobile at the lower water to cement ratios. Under the experimental conditions considered, the reduction in diffusive migration rates of both ^{137}Cs and ^{85}Sr in cement-based grouts resulting from a decrease in the water to solids ratio was comparable to the impact on the potential migration rates due to the addition of silica fume alone.

5.0 ACKNOWLEDGEMENTS

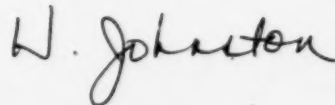
The authors wish to acknowledge the support of the Nuclear Engineering Department of Ontario Hydro for this study and the Concrete Technology Section of the Civil Research Department for the preparation of the grout specimens.

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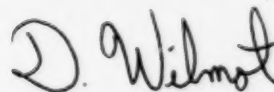


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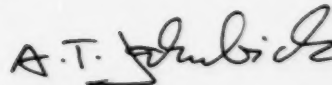


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APPENDIX A: Physical Measurements of Diffusion Samples

PHYSICAL CHARACTERISTICS OF GRANITE DIFFUSION CORE SLICES

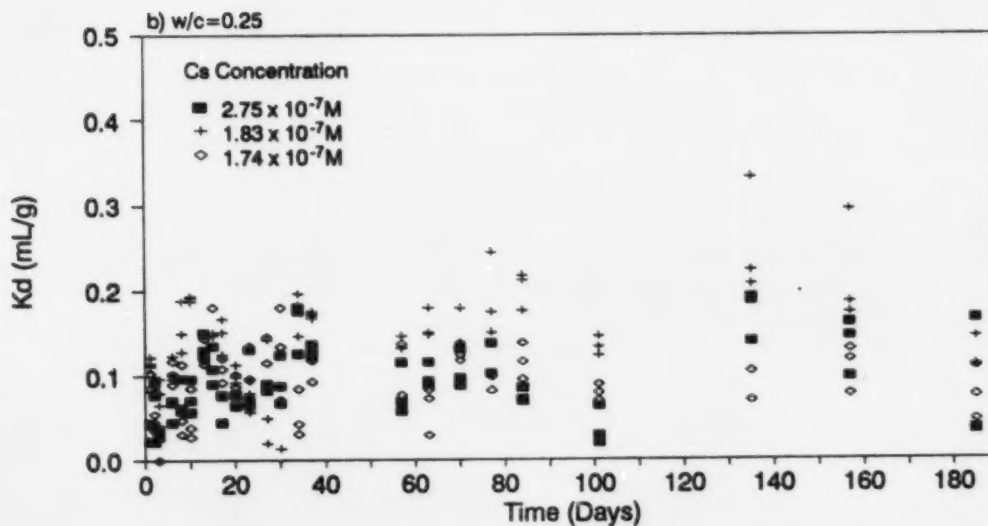
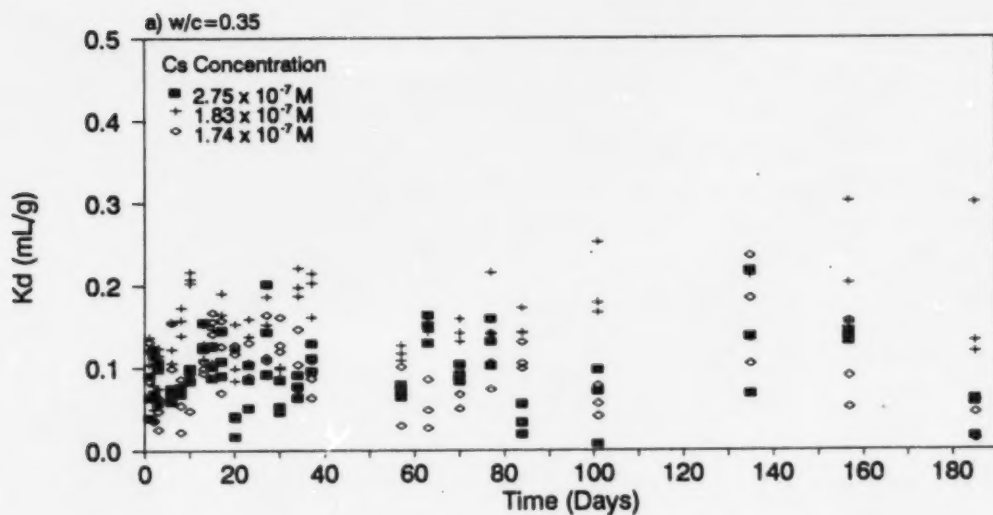
REP #	AVERAGE THICKNESS (mm)	AVERAGE DIAMETER (mm)	SURFACE AREA (mm ²)	SURFACE AREA(Total) (mm ²)	TOTAL VOLUME (mm ³)	TOTAL VOLUME (cc)
1	4.03	51.07	2048.44	4743.45	8255.20	8.26
2	4.02	51.08	2049.24	4743.58	8237.94	8.24
3	4.01	51.07	2048.44	4740.24	8214.23	8.21

PHYSICAL CHARACTERISTICS OF GROUT DIFFUSION DISCS - SCSS SOLUTION

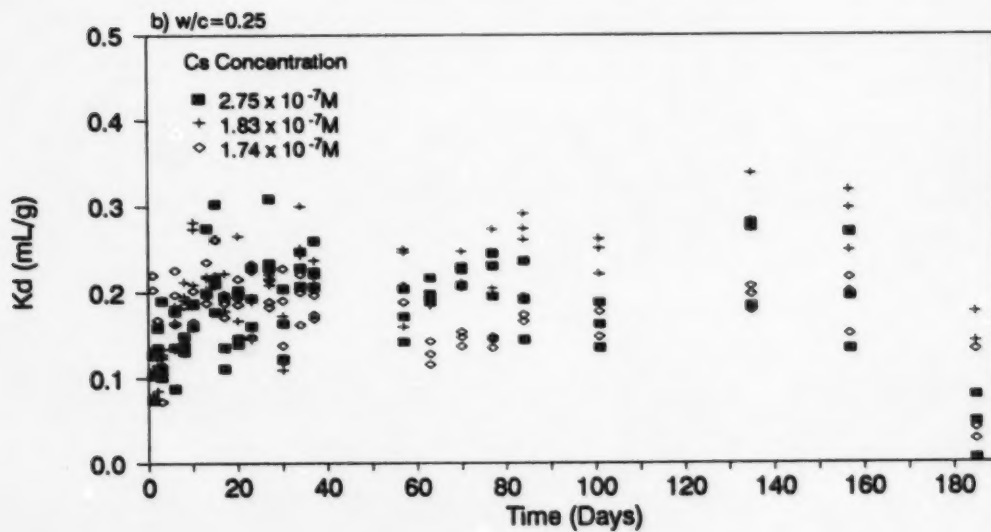
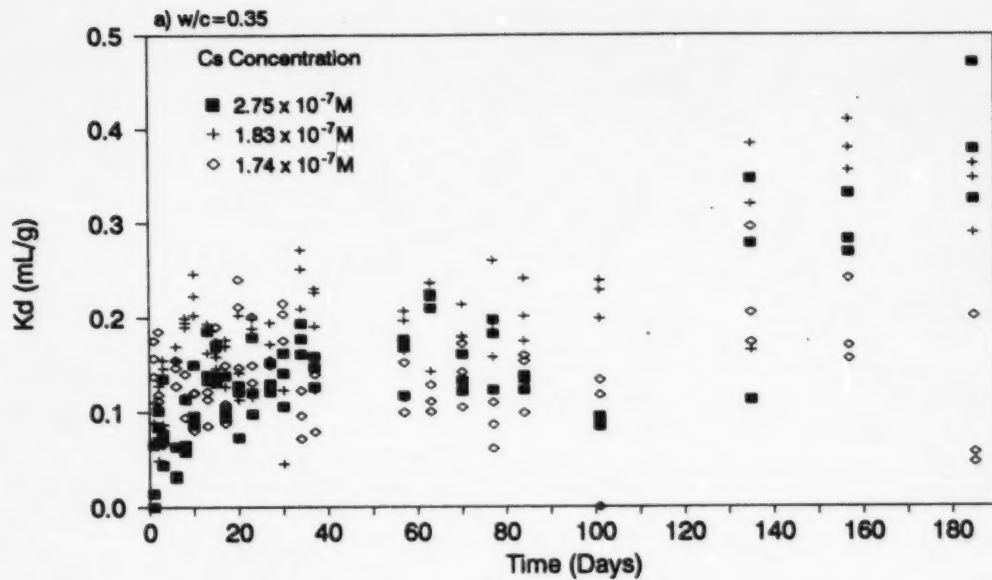
MIX	RATIO w/c	REP #	AVERAGE THICKNESS (mm)	AVERAGE DIAMETER (mm)	SURFACE AREA (mm ²)	SURFACE AREA(Total) (mm ²)	TOTAL VOLUME (mm ³)	TOTAL VOLUME (cc)
100%T50	0.35	1	5.89	50.89	2034.02	5009.72	11980.39	11.98
		2	5.88	50.90	2034.82	5009.90	11964.75	11.96
	0.25	1	5.88	50.99	2042.02	5025.97	12007.10	12.01
		2	5.87	51.01	2043.63	5027.94	11996.09	12.00
90%T50 10%SiF	0.35	1	5.85	51.02	2044.43	5026.52	11959.90	11.96
		2	5.88	51.06	2047.63	5038.48	12040.09	12.04
	0.25	1	5.89	50.95	2038.82	5020.42	12008.66	12.01
		2	5.87	50.94	2038.02	5015.44	11963.19	11.96
85%T50 15%SiF	0.35	1	5.90	50.62	2012.50	4963.26	11873.73	11.87
		2	5.88	50.60	2010.91	4956.53	11824.13	11.82
	0.25	1	5.90	51.10	2050.84	5048.85	12099.98	12.10
		2	5.91	51.07	2048.44	5045.08	12106.26	12.11

APPENDIX B: Cesium and Strontium Batch Test Results

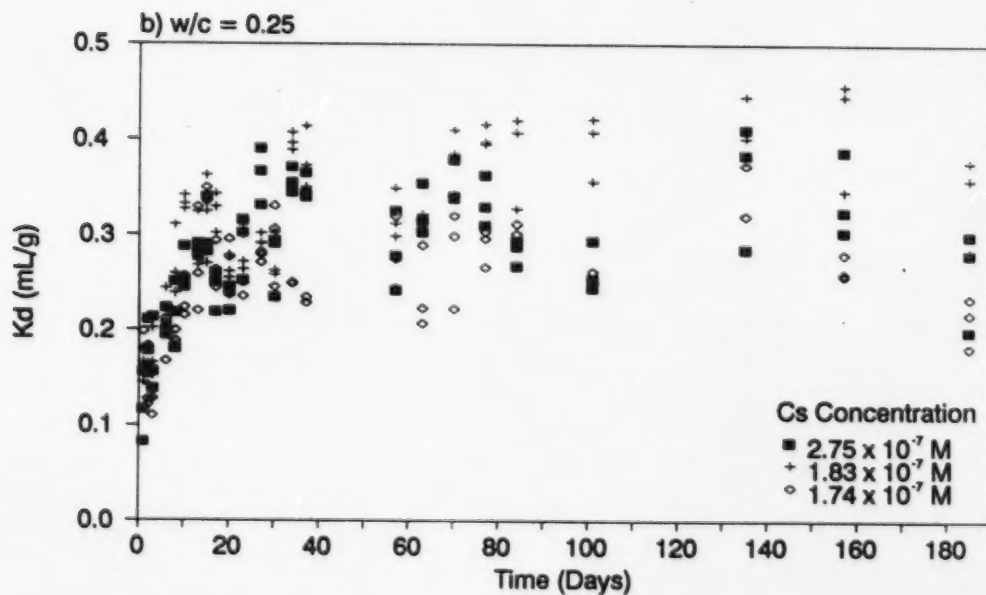
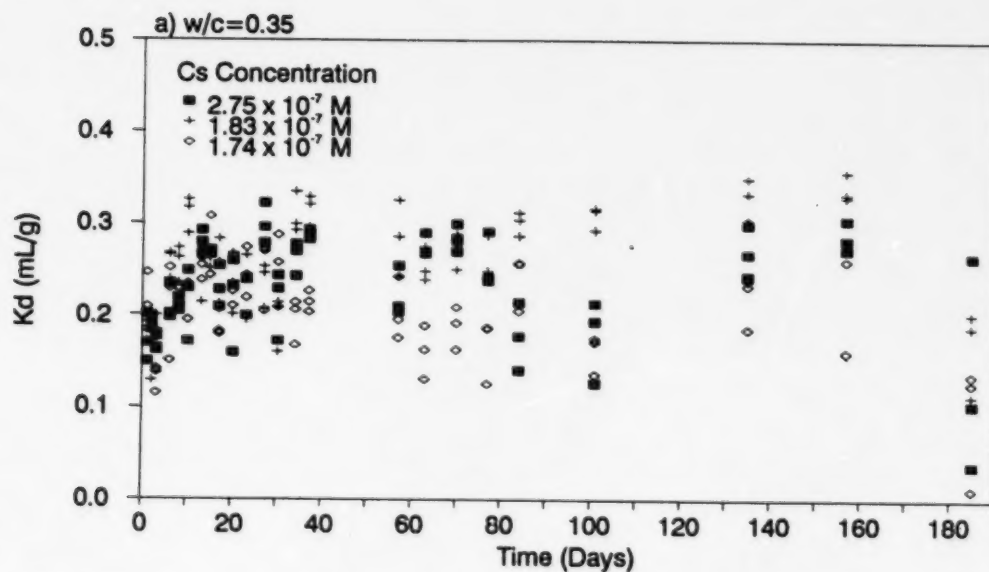
Cs Kd Results for 100% T50 Grout in SCSS



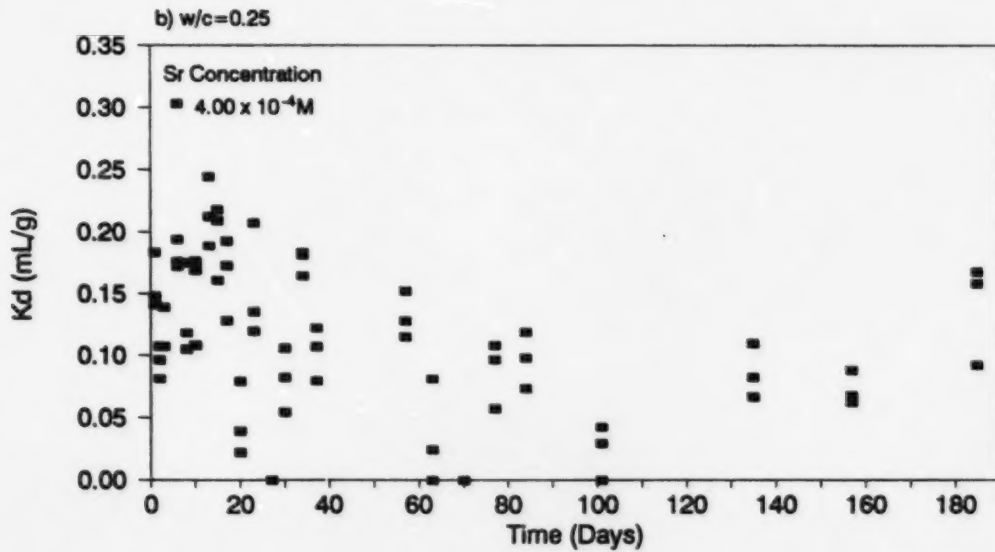
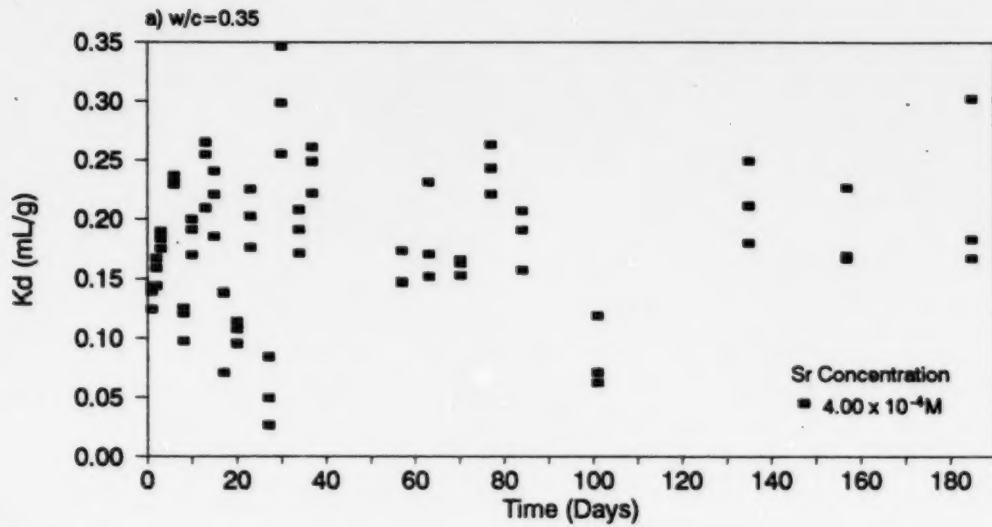
Cs Kd Results for 90% T50-10% SF Grout in SCSS



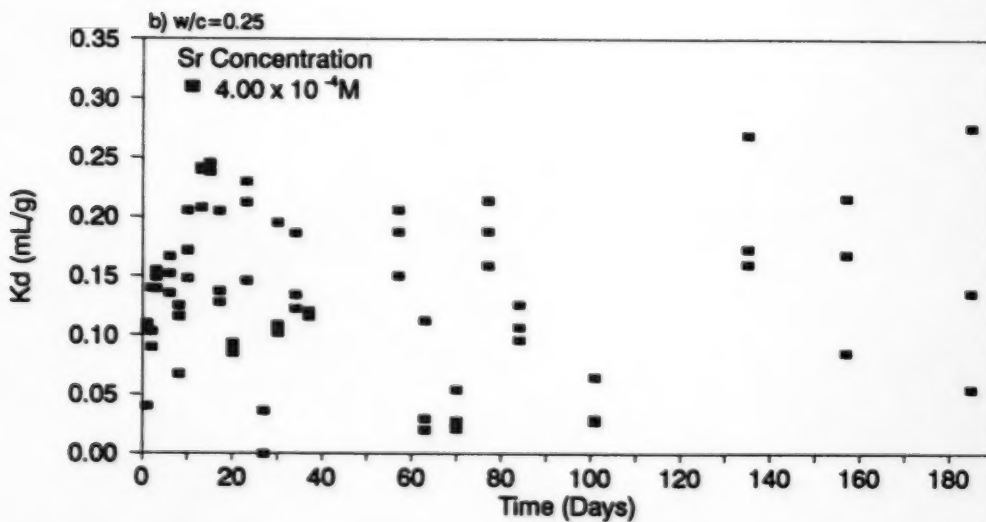
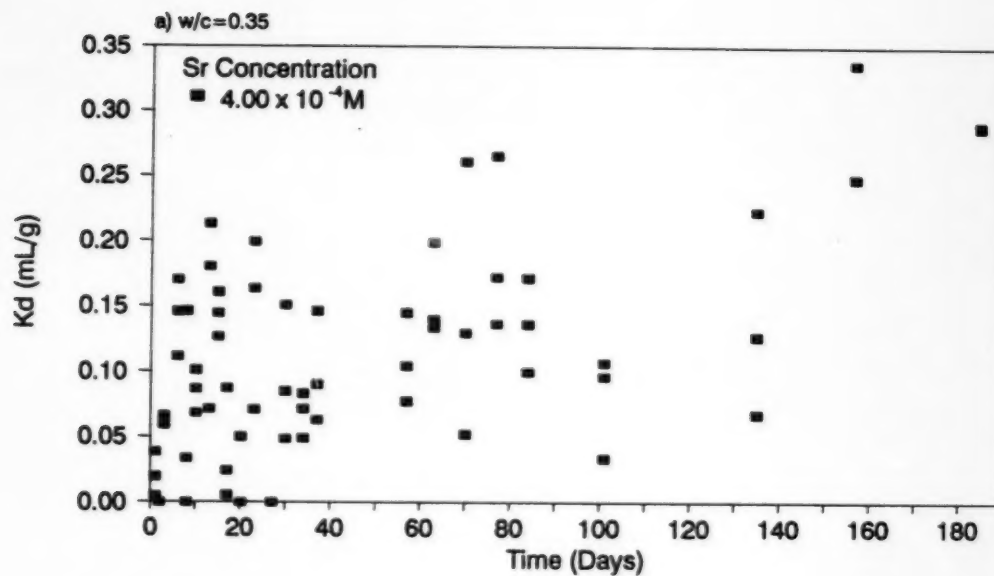
Cs Kd Results for 85% T50-15% SF Grout in SCSS



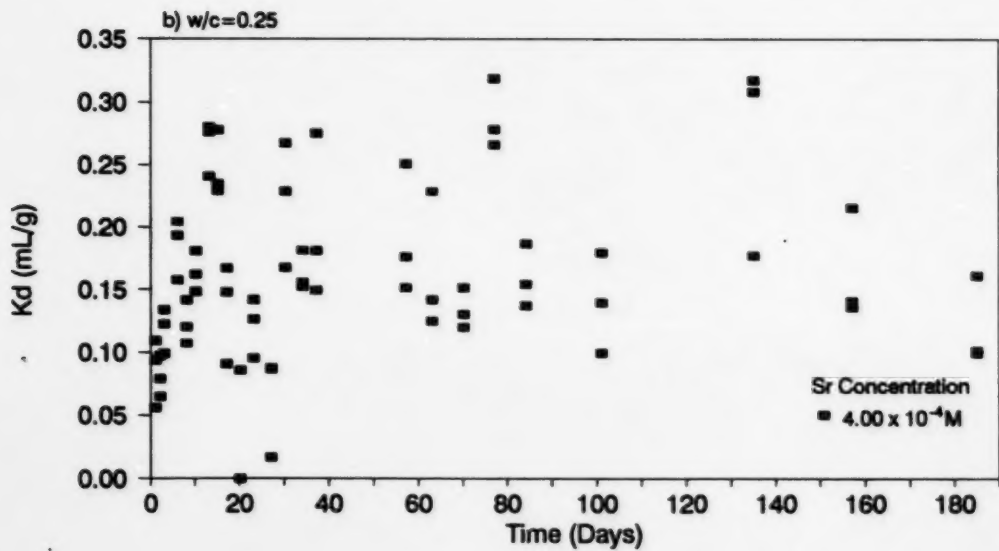
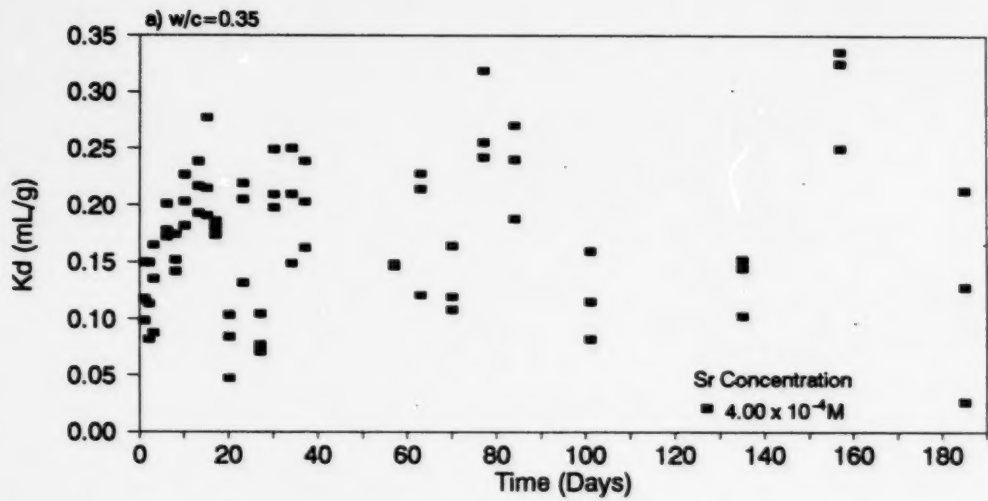
Sr Kd Results for 100% T50 Grout in SCSS



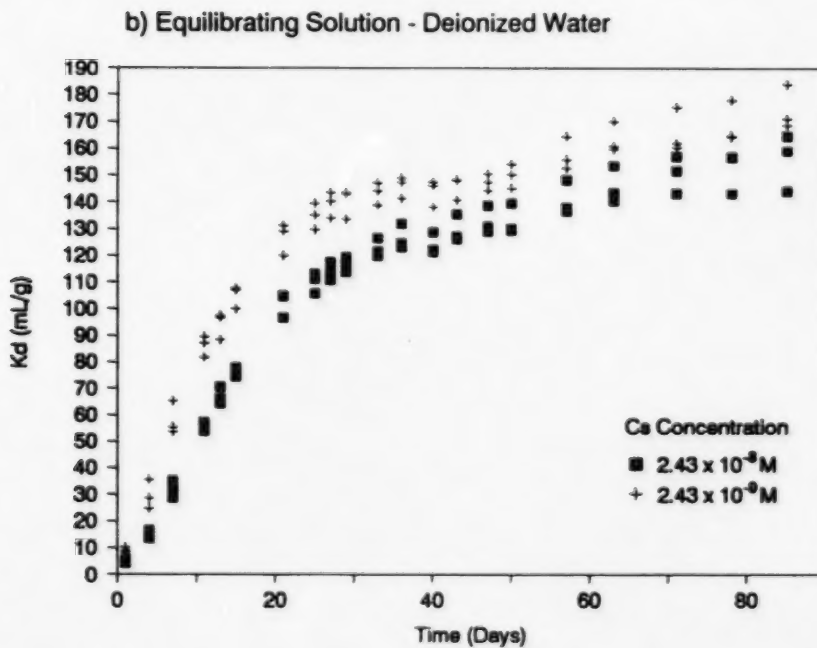
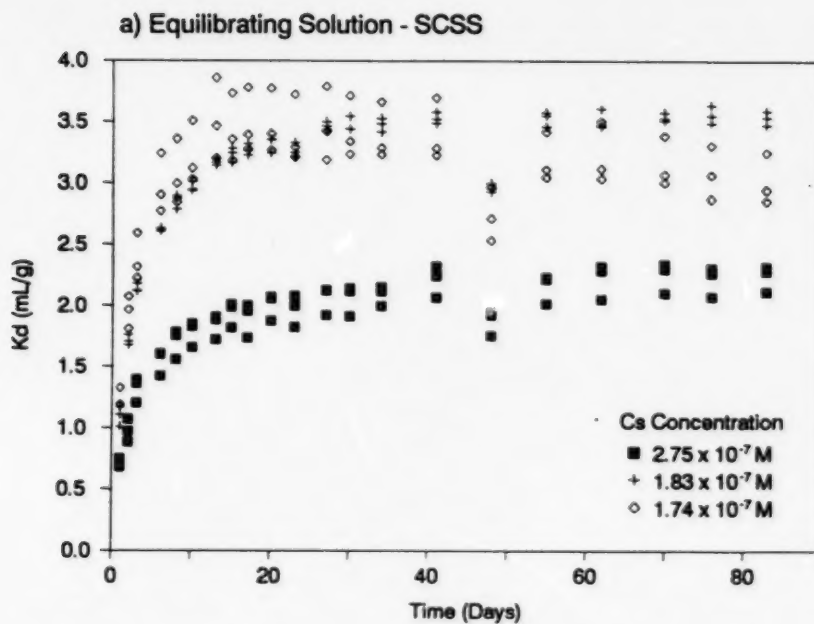
Sr Kd Results for 90% T50-10% SF Grout in SCSS



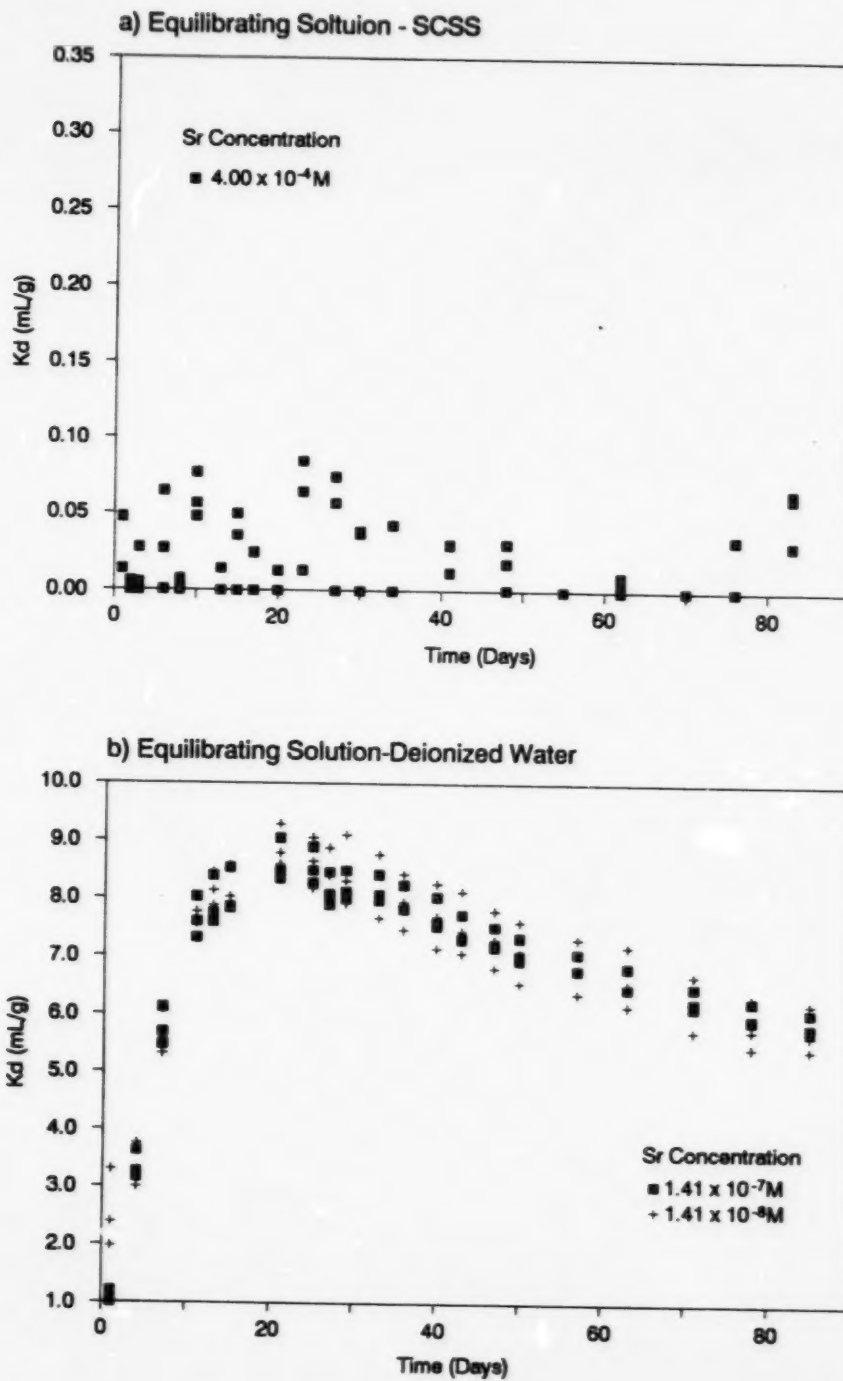
Sr Kd Results for 85% T50-15% SF Grout in SCSS



Granite Rock Core Cs Kd Results



Granite Rock Core Sr Kd Results



APPENDIX C: Tritium and ^{36}Cl Diffusion Results

Diffusion Results for Grout Mixes Using SCSS

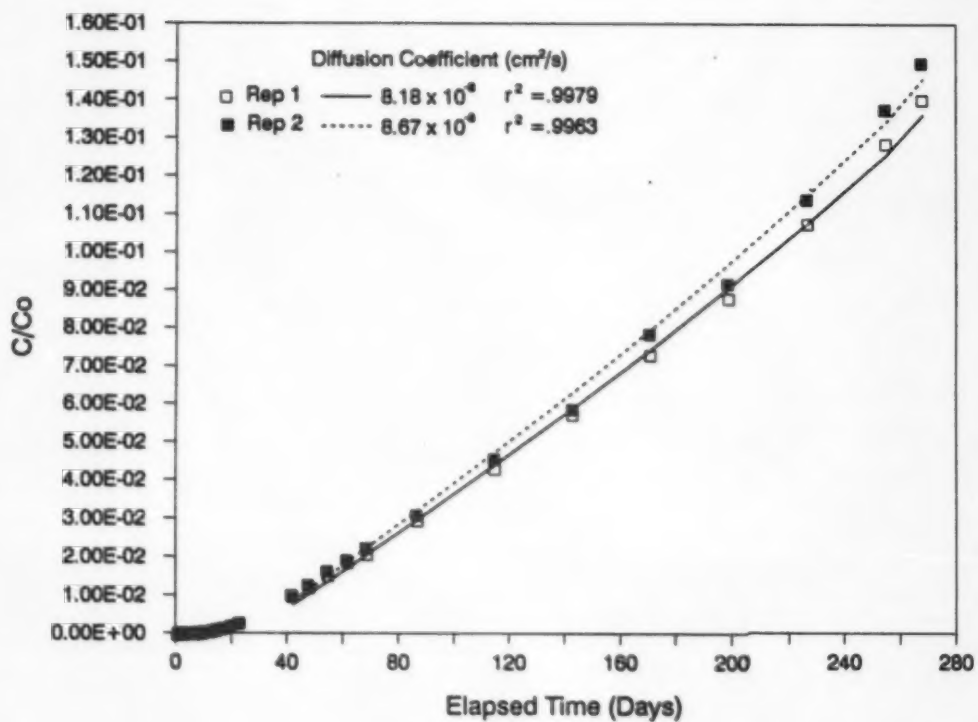
	Mix	Water to Cement Ratio	Replicate	Regression Line (# Data Points)	r ²	D x 10 ⁸ (cm ² /s)
H	100% T50	0.35	1	0.001276X - 0.03010 (13)	0.9979	8.18
			2	0.001356X - 0.03108 (13)	0.9963	8.67
		0.25	1	0.001096X - 0.02391 (13)	0.9992	11.24
			2	0.000684X - 0.01810 (13)	0.9992	7.01
	90% T50 10% SF	0.35	1	0.001013X - 0.05769 (7)	0.9961	5.20
			2	0.001048X - 0.06076 (7)	0.9927	5.38
		0.25	1	0.000708X - 0.04636 (6)	0.9979	5.77
			2	0.000743X - 0.04494 (7)	0.9986	6.06
	85%T50 15% SF	0.35	1	0.000796X - 0.06023 (6)	0.9961	4.65
			2	0.000793X - 0.06046 (6)	0.9973	4.64
		0.25	1	0.000550X - 0.02359 (16)	0.9920	4.90
			2	0.0005397X - 0.02359 (16)	0.9871	4.81
36Cl	100% T50	0.35	1	0.001078X - 0.04122 (13)	0.9953	6.91
			2	0.001163X - 0.04367 (13)	0.9936	7.46
		0.25	1	0.000668X - 0.04481 (7)	0.9958	6.84
			2	0.000343X - 0.02761 (7)	0.9955	3.51
	90% T50 10% SF	0.35	1	0.00059X - 0.06302 (6)	0.9904	3.04
			2	0.000719X - 0.08925 (4)	0.9929	3.70
		0.25	1	0.000173X - 0.02828 (4)	0.9939	1.41
			2	0.0002301X - 0.03163 (4)	0.9969	1.64
	85% T50 15% SF	0.35	1	0.000282X - 0.04409 (4)	0.9934	1.65
			2	0.000260X - 0.04058 (4)	0.9946	1.52
		0.25	1	0.000282X - 0.0986 (8)	0.9987	2.52
			2	0.000282X - 0.10100 (7)	0.9990	2.52

Time Lag of Grout Diffusion Results Using SCSS

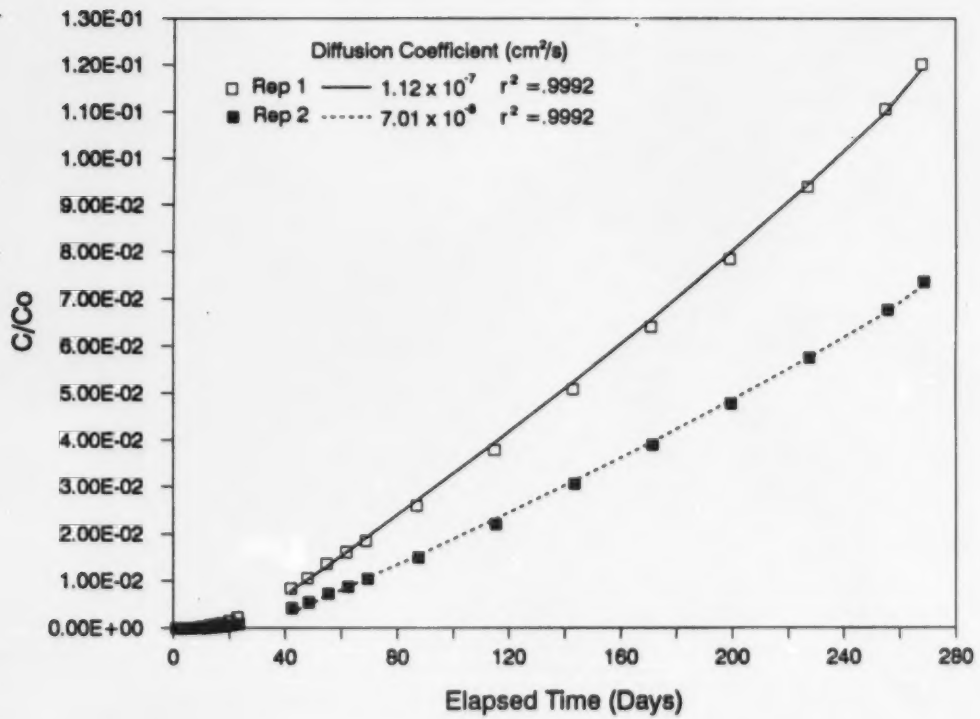
	Mix	Water to Cement Ratio	Replicate	D x10 ⁸ (cm ² /s)	Time Lag (days)	
					Predicted (Kd=0)	Observed
³ H	100% T50	0.35	1	8.18	8.2	24
			2	8.67	7.7	23
		0.25	1	11.24	5.9	22
			2	7.01	9.5	27
	90% T50 10% SF	0.35	1	5.20	13	57
			2	5.38	12	28
		0.25	1	5.77	12	65
			2	6.06	11	61
	85%T50 15% SF	0.35	1	4.65	14	76
			2	4.64	14	82
		0.25	1	4.90	14	43
			2	4.81	13	44
³⁶ Cl	100% T50	0.35	1	6.91	9.7	38
			2	7.46	8.9	37
		0.25	1	6.84	9.7	67
			2	3.51	19	81
	90% T50 10% SF	0.35	1	3.04	22	107
			2	3.70	18	124
		0.25	1	1.41	48	164
			2	1.64	41	138
	85% T50 15% SF	0.35	1	1.65	41	156
			2	1.52	44	156
		0.25	1	2.52	27	351
			2	2.52	27	359

TRITIUM DIFFUSION IN SCSS

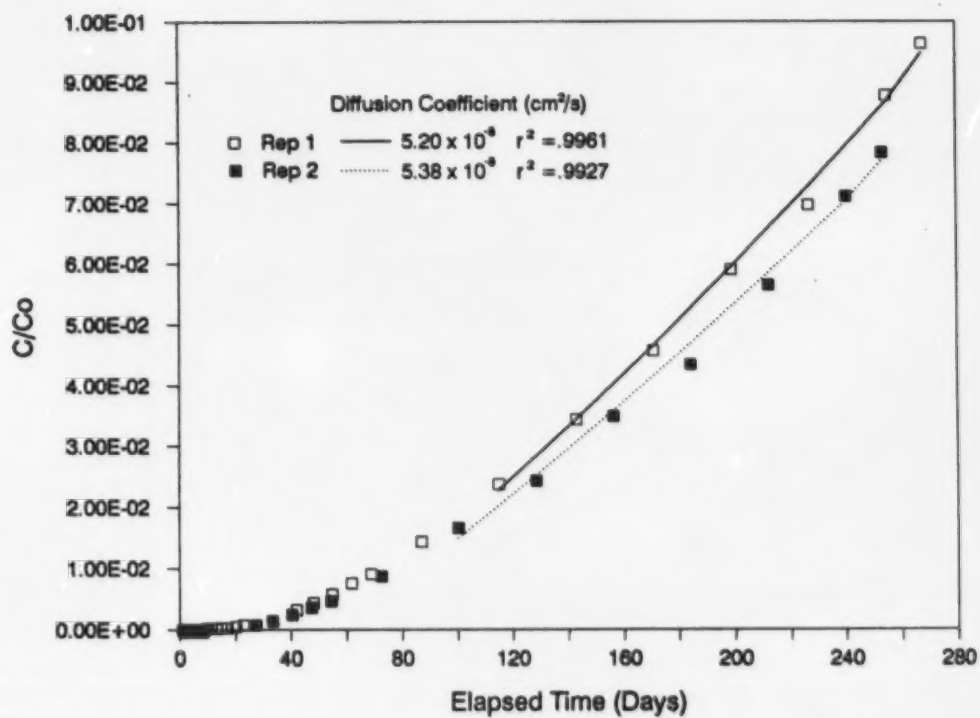
100% T50 CEMENT W/C=.35



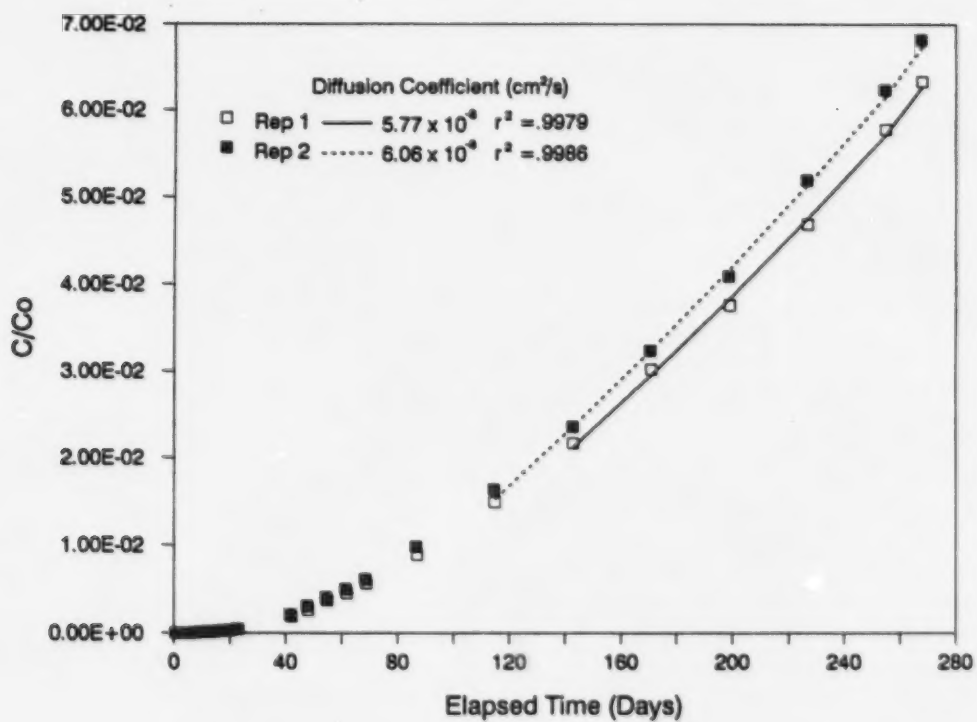
TRITIUM DIFFUSION IN SCSS
100% T50 CEMENT W/C=.25



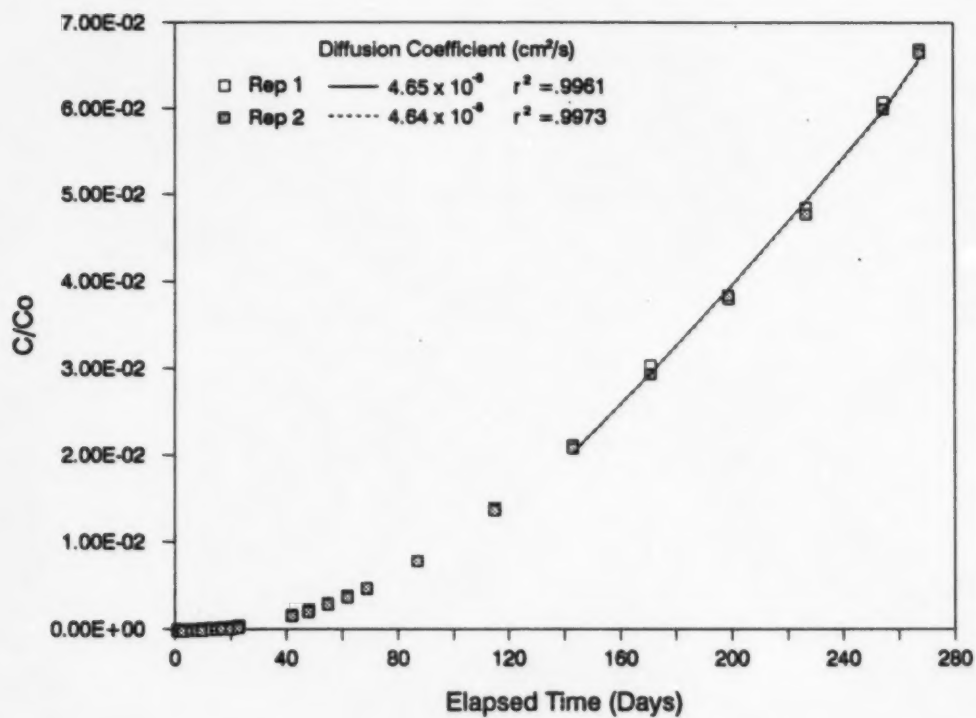
TRITIUM DIFFUSION IN SCSS
90% T50 CEMENT:10% SILICA FUME W/C=.35



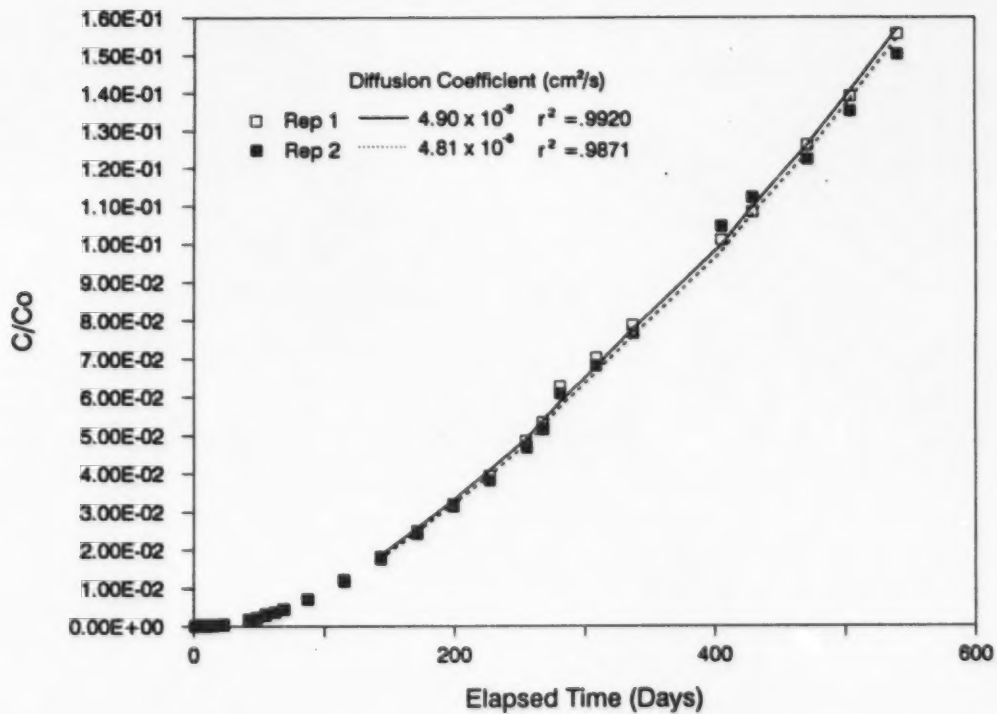
TRITIUM DIFFUSION IN SCSS
90% T50 CEMENT:10% SILICA FUME W/C=.25



TRITIUM DIFFUSION IN SCSS
85% T50 CEMENT:15% SILICA FUME W/C=.35

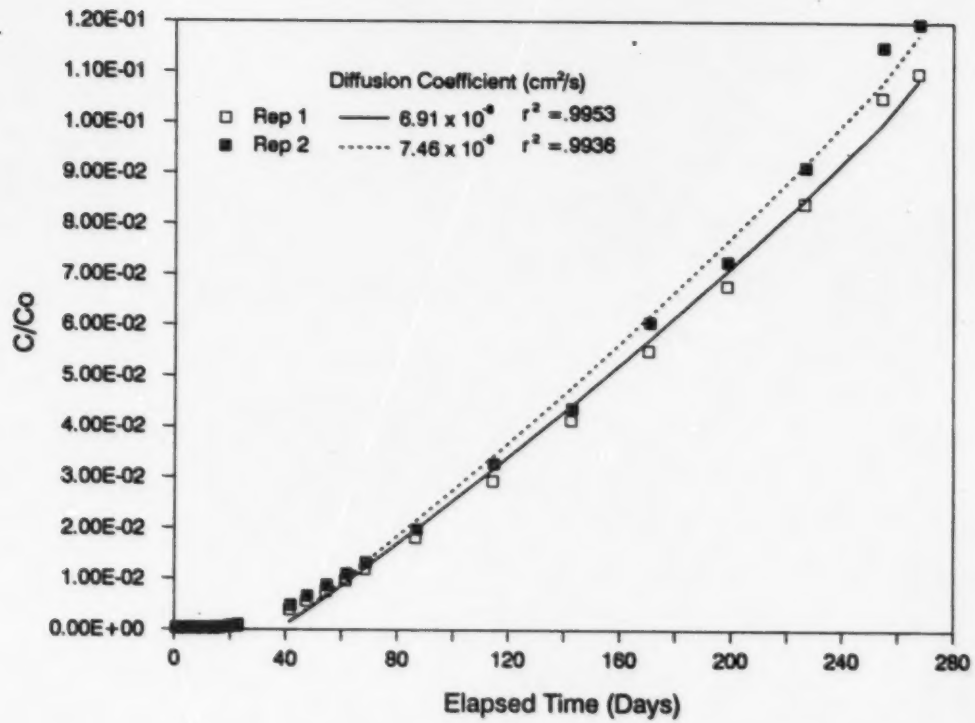


TRITIUM DIFFUSION IN SCSS
85% T50 CEMENT:15% SILICA FUME W/C=.25



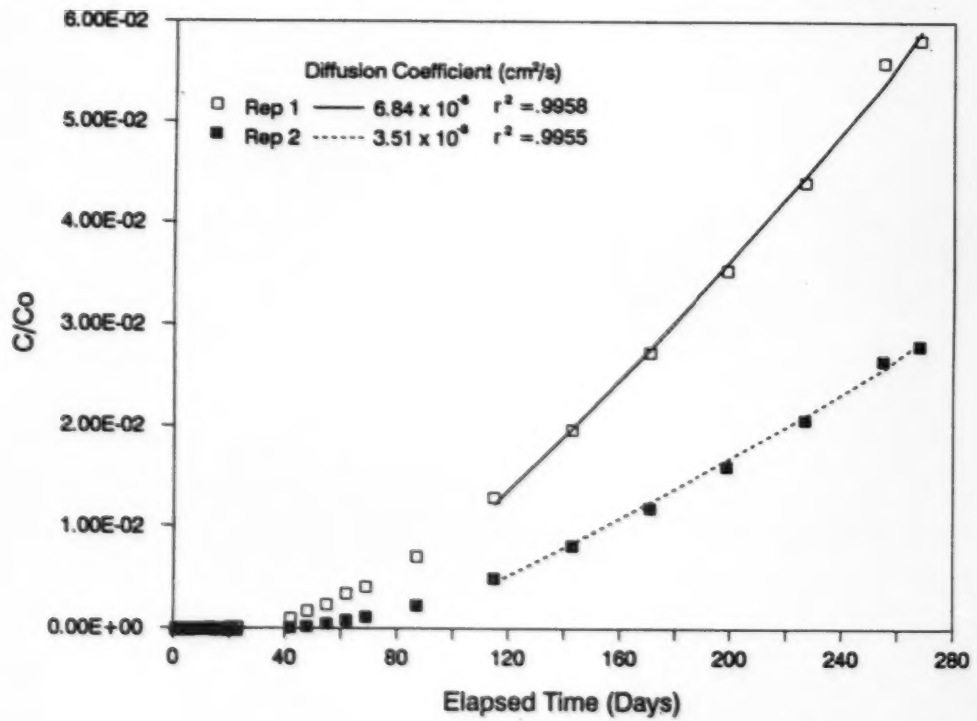
CHLORIDE DIFFUSION IN SCSS

100% T50 CEMENT W/C=.35

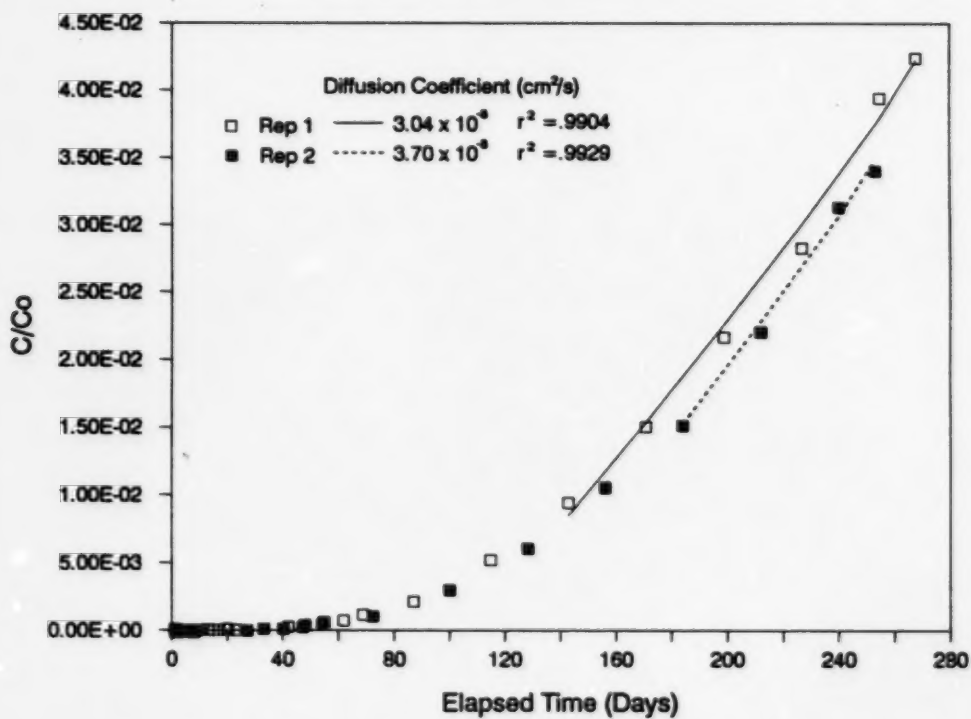


CHLORIDE DIFFUSION IN SCSS

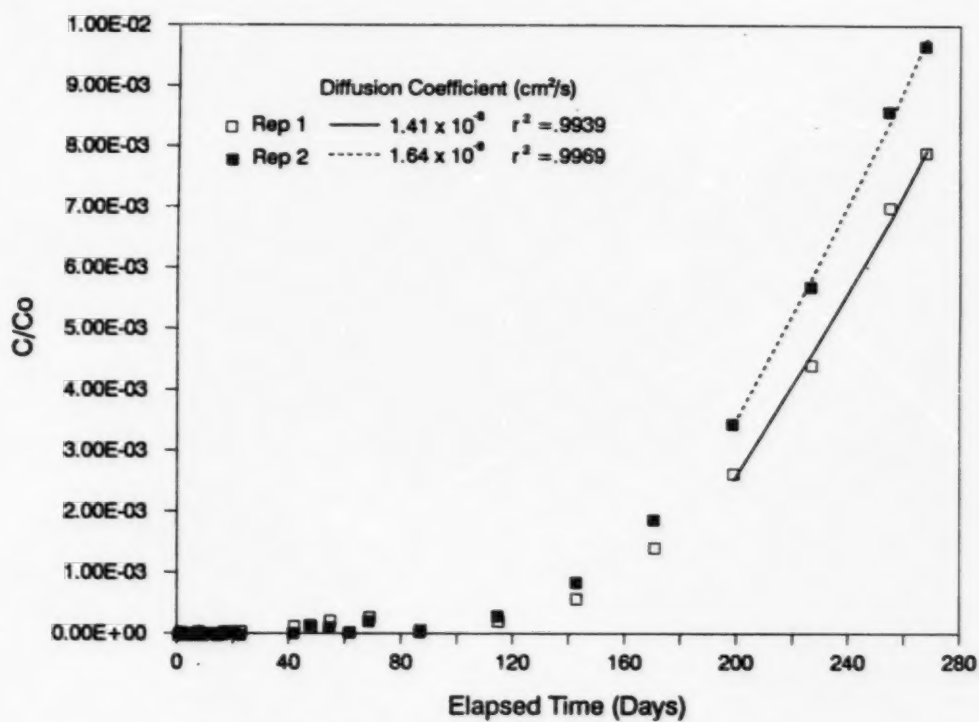
100% T50 CEMENT W/C=.25



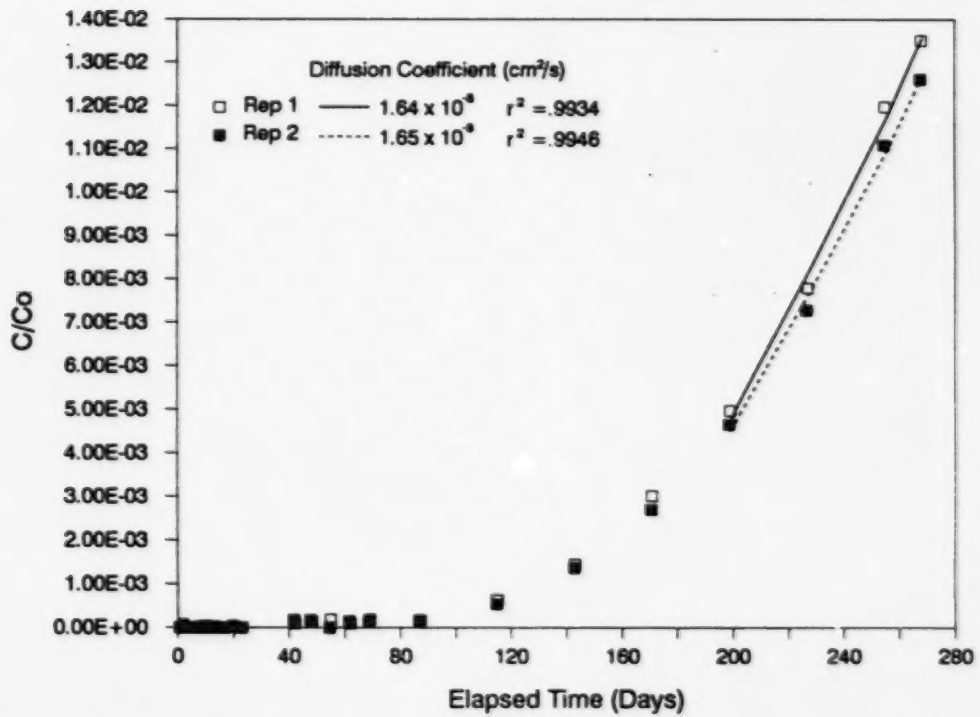
CHLORIDE DIFFUSION IN SCSS
 90% T50 CEMENT:10% SILICA FUME W/C=.35



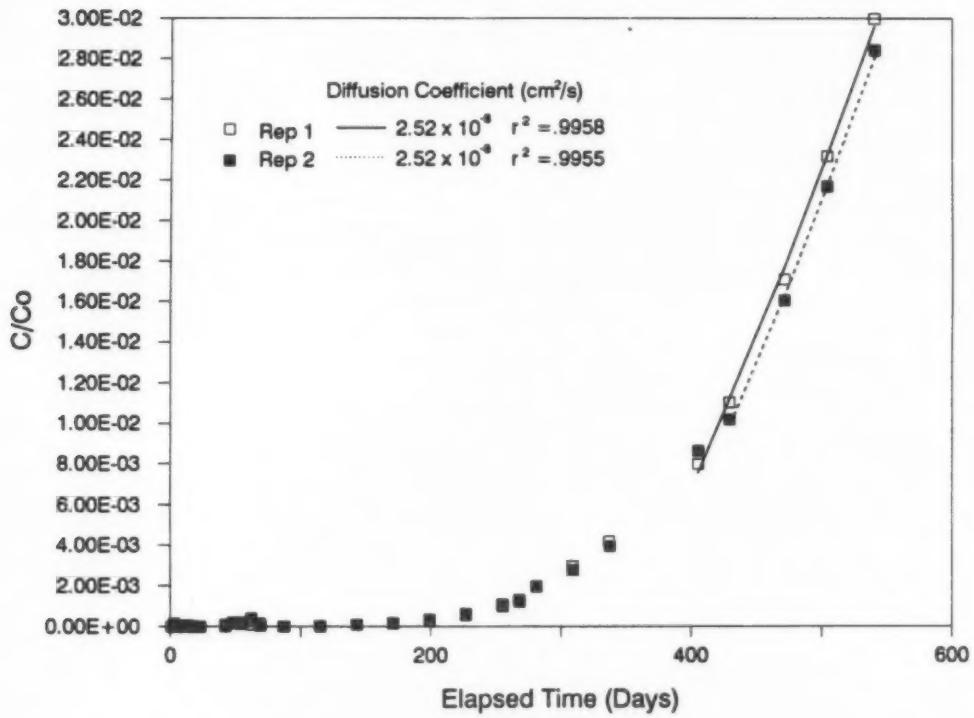
CHLORIDE DIFFUSION IN SCSS
90% T50 CEMENT:10% SILICA FUME W/C=.25



CHLORIDE DIFFUSION IN SCSS
85% T50 CEMENT:15% SILICA FUME W/C=.35



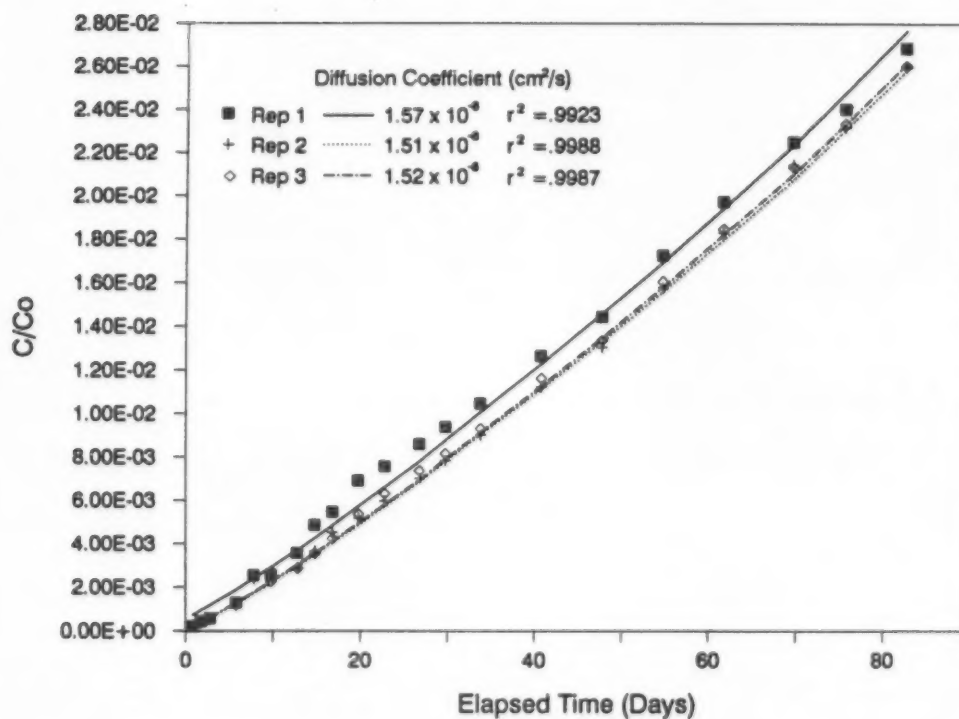
CHLORIDE DIFFUSION IN SCSS
85% T50 CEMENT:15% SILICA FUME W/C = .25



Diffusion Results for Lac du Bonne Granite in SCSS

	Isotope	Replicate	Regression Line*	r ²	D x 10 ⁶ (cm ² /s)
Lac du Bonne Granite	³ H	1	0.000542X+0.001348	0.9923	1.57
		2	0.000521X+0.000095	0.9988	1.51
		3	0.000525X+0.000151	0.9987	1.52
	³⁶ Cl	1	0.000351X+0.001331	0.9824	1.01
		2	0.000332X-0.00001	0.9984	0.96
		3	0.000341X+0.000099	0.9989	0.99
* CVI/CoA versus t					

TRITIUM DIFFUSION RESULTS FOR GRANITE IN SCSS



CHLORIDE DIFFUSION RESULTS FOR GRANITE IN SCSS

